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**WO 01/70868 A1**

(54) Title: STABILIZING MIXTURES FOR ORGANIC POLYMERS

(57) Abstract: Stabilizing mixtures for organic polymers comprising: (a) at least one compound belonging to the group of 3-pyrazolidinones; (b) at least one compound belonging to the group of organic phosphites or phosphonites; (c) at least one compound belonging to the group of sterically hindered phenols; and, optionally, (d) at least one compound belonging to the group of sterically hindered amines. The above mixtures can be used as stabilizers of organic polymers to degradation caused by oxygen, heat and/or light.

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## STABILIZING MIXTURES FOR ORGANIC POLYMERS

The present invention relates to stabilizing mix-  
tures for organic polymers.

10 More specifically, the present invention relates to  
stabilizing mixtures for organic polymers comprising at  
least one compound belonging to the group of 3-  
pyrazolidinones, at least one compound belonging to the  
group of organic phosphites or phosphonites, at least one  
15 compound belonging to the group of sterically hindered  
phenols and, optionally, at least one compound belonging  
to the group of sterically hindered amines and their use  
in the stabilization of organic polymers to degradation  
caused by oxygen, heat and/or light.

20 The present invention also relates to the polymeric  
compositions stabilized with the above stabilizing mix-  
tures and the end-products obtained by their processing.

Japanese patent application JP 56/086165 describes  
the use of derivatives of 3-pyrazolidinone as colour sta-  
25 bilizers and antioxidants in the photographic field.

Organic phosphites, organic phosphonites and phosphoramides are known in the art as co-stabilizers, secondary antioxidants and process stabilizers for organic polymers among which polyolefins. Examples of these compounds can be found, for example, in R. Gächter/H. Müller (Ed.), "Plastic Additives Handbook" (1990), 3<sup>rd</sup> Ed., page 47, Hanser, München.

U.S. patent 4,360,617 describes stabilizing mixtures comprising symmetrical triarylphosphites and phenolic antioxidants, particularly useful in the stabilization to degradation caused by oxygen, heat and/or light, of various organic polymers such as, for example, polyurethanes, polyacrylonitrile, polyamide 12 or polystyrene.

Sterically hindered amines, in particular those carrying 2,2,6,6-tetramethylpiperidine groups in the molecule, are known as HALS.

The known stabilizers, however, are not capable of completely satisfying all the problems to be solved by a stabilizer such as, for example, storage life, water absorption, sensitivity to hydrolysis, stabilization during the polymer processing, colour properties of the stabilized polymer, volatility, migration within the stabilized polymer, compatibility with the polymer to be stabilized and improvement in light protection. There is consequently a continuous need for stabilizers for or-

ganic polymers capable of providing a better stabilization thereof.

The Applicant has now found that stabilizing mixtures comprising at least one compound belonging to the group of 3-pyrazolidinones, at least one compound belonging to the group of organic phosphites or phosphonites, and, optionally, at least one compound belonging to the group of sterically hindered phenols and, optionally, at least one compound belonging to the group of sterically hindered amines, are capable of providing a better stabilization to degradation caused by oxygen, heat and/or light, of the organic polymers to which they are added.

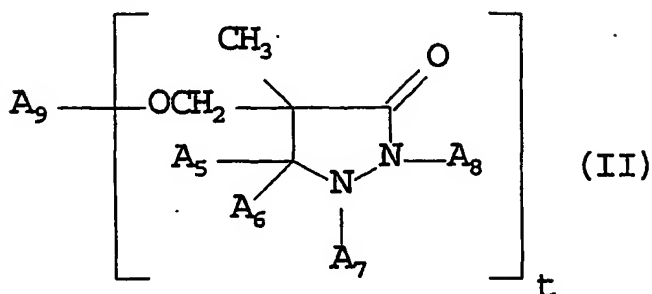
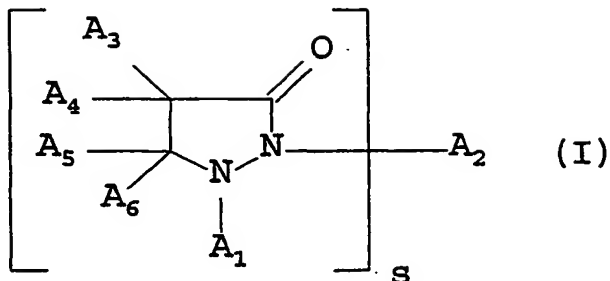
An object of the present invention therefore relates to stabilizing mixtures for organic polymers comprising:

- (a) at least one compound belonging to the group of 3-pyrazolidinones;
- (b) at least one compound belonging to the group of organic phosphites or phosphonites; and, optionally,
- (c) at least one compound belonging to the group of sterically hindered phenols; and, optionally,
- (d) at least one compound belonging to the group of sterically hindered amines.

Compounds belonging to the group of 3-pyrazolidinones (a) useful for the purposes of the present invention are selected from those having general for-



mula (I) or (II):

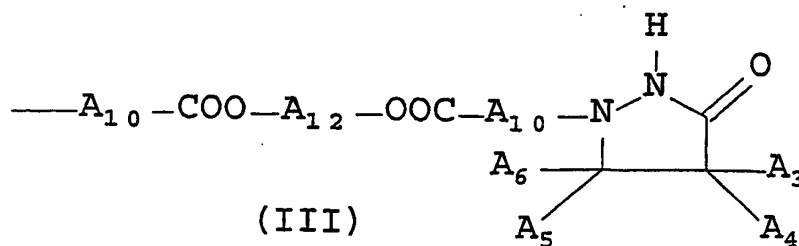


wherein:

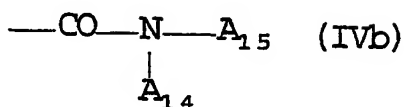
- A<sub>1</sub> represents a linear or branched C<sub>1</sub>-C<sub>18</sub> alkyl group; a C<sub>5</sub>-C<sub>12</sub> cycloalkyl group, said cycloalkyl group optionally mono-, di- or tri-substituted with a linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl group; a phenyl group, said phenyl group optionally mono-, di- or tri-substituted with a linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl or alkoxy group, and/or mono-substituted with a hydroxyl group; a C<sub>7</sub>-C<sub>9</sub> phenylalkyl group, said phenylalkyl group optionally mono-, di- or tri-substituted on the phenyl with a linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl group, and/or mono-substituted with a hydroxyl group; an -A<sub>10</sub>-COOA<sub>11</sub> wherein A<sub>10</sub> represents a linear or branched C<sub>1</sub>-C<sub>6</sub> alkylene group and A<sub>11</sub>

represents a linear or branched C<sub>1</sub>-C<sub>18</sub> alkyl group, a C<sub>5</sub>-C<sub>12</sub> cycloalkyl group, said cycloalkyl group optionally mono-, di- or tri-substituted with a linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl group, a linear or branched C<sub>3</sub>-C<sub>18</sub> alkenyl group, a C<sub>7</sub>-C<sub>9</sub> phenylalkyl group, said phenylalkyl group optionally mono-, di- or tri-substituted on the phenyl with a linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl group;

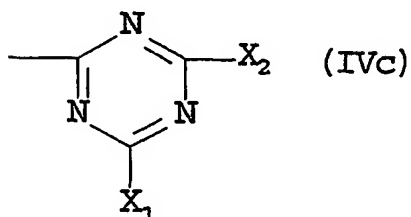
- A<sub>3</sub>, A<sub>4</sub>, A<sub>5</sub> and A<sub>6</sub>, the same or different, represent a hydrogen atom; a linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl group; a phenyl group;
- s is 1 or 2;
- when s is 1, A<sub>2</sub> represents a hydrogen atom; a linear or branched C<sub>1</sub>-C<sub>18</sub> alkyl group; a linear or branched C<sub>3</sub>-C<sub>6</sub> alkenyl group; a C<sub>7</sub>-C<sub>9</sub> phenylalkyl group, said phenylalkyl group optionally mono-, di- or tri-substituted on the phenyl with a linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl group; an -A<sub>10</sub>-COOA<sub>11</sub> wherein A<sub>10</sub> and A<sub>11</sub> have the same meanings defined above; and, when A<sub>2</sub> represents a hydrogen atom, A<sub>1</sub> may also represent a group having general formula (III):



- wherein  $A_3$ ,  $A_4$ ,  $A_5$ ,  $A_6$  and  $A_{10}$  have the same meanings defined above and  $A_{12}$  represents a linear or branched  $C_2$ - $C_{12}$  alkylene group; a  $C_4$ - $C_{12}$  alkylene group interrupted by 1, 2 or 3 oxygen atoms; a cyclohexylene group; a cyclohexylenedimethylene group; an isopropylidenedicyclohexylidene group;
- when  $s$  is 2,  $A_2$  represents a linear or branched  $C_2$ - $C_{12}$  alkylene group, or a xylylene group;
- $A_7$  represents a phenyl group optionally mono-, di- or tri-substituted with a linear or branched  $C_1$ - $C_4$  alkyl or alkoxy group, and/or mono-substituted with a hydroxyl group;
- $A_8$  represents a hydrogen atom; a linear or branched  $C_1$ - $C_{18}$  alkyl group; a linear or branched  $C_3$ - $C_6$  alkenyl group; a  $C_7$ - $C_9$  phenylalkyl group, said phenylalkyl group optionally mono-, di-, or tri-substituted on the phenyl with a linear or branched  $C_1$ - $C_4$  alkyl group; an  $-A_{10}-COOA_{11}$  group wherein  $A_{10}$  and  $A_{11}$  have the same meanings defined above;
- $t$  is 1, 2, 3 or 4;
- when  $t$  is 1,  $A_9$  represents an acyl-aliphatic, cycloaliphatic, aromatic, arylaliphatic or heterocyclic group containing not more than 22 carbon atoms; or one of the following groups having general formula (IVa)-(IVc):

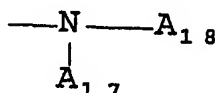


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wherein  $A_{13}$  has the same meanings as  $A_{11}$  described above;  $A_{14}$  and  $A_{15}$ , the same or different, have the same meanings as  $A_{11}$  described above, or they represent a hydrogen atom, or a phenyl group, or  $A_{14}$  and  $A_{15}$  considered jointly with the nitrogen atom to which they are bound, represent a heterocyclic group with 5-7 atoms;  $X_1$  and  $X_2$ , the same or different, represent an  $-\text{O}-A_{16}$  group, or a group having the formula:

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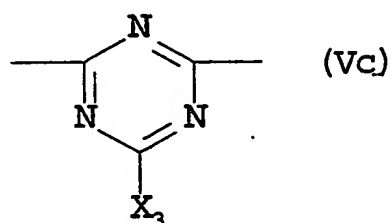
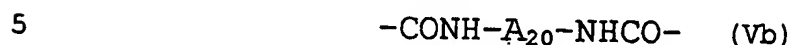
wherein  $A_{16}$ ,  $A_{17}$  and  $A_{18}$ , the same or different, have the same meanings as  $A_{14}$  and  $A_{15}$  described above, or  $A_{17}$  and  $A_{18}$  considered jointly with the nitrogen atom to which they are bound, represent a heterocyclic group with 5-7 atoms;

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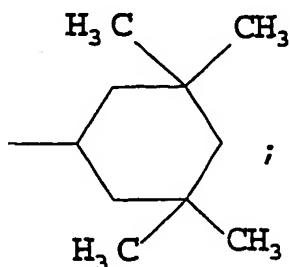
- when  $t$  is 2,  $A_9$  represents a diacyl-aliphatic, cycloaliphatic, aromatic, arylaliphatic or heterocy-

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clic group containing not more than 22 carbon atoms;  
or one of the following groups having general formula (Va)-(Vc):



wherein  $\text{A}_{19}$  represents a linear or branched  $\text{C}_2$ - $\text{C}_{12}$  alkylene group, a  $\text{C}_4$ - $\text{C}_{12}$  alkylene group interrupted by 1, 2 or 3 oxygen atoms, a cyclohexylene group, a cyclohexylenedimethylene group, an isopropylidenedi-cyclohexylidene group, a phenylene group, a xylylene group, an isopropylidenediphenylene group;  $\text{A}_{20}$  represents a linear or branched  $\text{C}_2$ - $\text{C}_{12}$  alkylene group, a cyclohexylene group, a cyclohexylenedimethylene group, a methylenedicyclohexylene group, a phenylene group, a methylphenylene group, a xylylene group, a methylenediphenylene group, an oxydiphenylene group, a group having the formula:



$X_3$  has the same meanings as  $X_1$  and  $X_2$  defined above;

- when  $t$  is 3,  $A_9$  represents a triacyl-aliphatic, aromatic or heterocyclic group containing not more than 18 carbon atoms, or a 2,4,6-triyl-1,3,5-triazine group;

- when  $n$  is 4,  $A_9$  represents a tetra-acyl-aliphatic or aromatic group containing not more than 18 carbon atoms.

Examples of linear or branched alkyl groups, having not more than 18 carbon atoms are: methyl, ethyl, propyl, isopropyl, butyl, 2-butyl, isobutyl, t-butyl, pentyl, 2-pentyl, hexyl, heptyl, octyl, 2-ethylhexyl, t-octyl, nonyl, decyl, undecyl, dodecyl, tridecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, etc.

Examples of linear or branched alkoxy groups, having not more than 18 carbon atoms are: methoxy, ethoxy, propoxy, isopropoxy, butoxy, isobutoxy, pentoxy, isopentoxy, hexoxy, heptoxy, octoxy, nonoxy, decyloxy, dodecyloxy, tetradecyloxy, hexadecyloxy, octadecyloxy, etc.

Examples of  $C_5$ - $C_{12}$  cycloalkyl groups, optionally mono-, di- or tri-substituted with a linear or branched  $C_1$ - $C_4$  alkyl group, are: cyclopentyl, methylcyclopentyl, dimethylcyclopentyl, cyclohexyl, methylcyclohexyl, dimethylcyclohexyl, trimethylcyclohexyl, t-butylcyclohexyl, cyclo-octyl, cyclodecyl, cyclododecyl, etc.

Examples of alkenyl groups having not more than 18 carbon atoms are: allyl, 2-methyallyl, butenyl, hexenyl, undecenyl, octadecenyl.

Examples of C<sub>7</sub>-C<sub>9</sub> phenylalkyl groups, optionally  
5 mono-, di-, or tri-substituted on the phenyl, are: benzyl, methylbenzyl, dimethylbenzyl, trimethylbenzyl, t-butylbenzyl, 2-phenylethyl, 3,5-di-t-butyl-4-hydroxybenzyl, etc.

Examples of substituted phenyl groups are: methyl-  
10 phenyl, dimethylphenyl, trimethylphenyl, 3,5-di-t-butyl-4-methylphenyl, methoxyphenyl, ethoxyphenyl, dimethoxyphenyl, trimethoxyphenyl, hydroxyphenyl, 3,5-di-t-butyl-4-hydroxyphenyl, etc.

Examples of heterocyclic groups with 5-7 atoms are:  
15 1-pyrrolidyl, 1-piperidyl, 4-morpholinyl, 4-methyl-1-piperazinyl, 1-hexahydroazepinyl, etc.

Examples of acyl-aliphatic, cycloaliphatic, aromatic, arylaliphatic or heterocyclic groups having not more than 22 carbon atoms are those deriving from the  
20 following acids: formic, acetic, propionic, butyric, isobutyric, valeric, pivalic, hexanoic, heptanoic, octanoic, 2-ethylhexanoic, nonanoic, decanoic, undecanoic, dodecanoic, tetradecanoic, hexadecanoic, octadecanoic, eicosanoic, docosanoic, acrylic, methacrylic, crotonic, undecenoic, octadecenoic, cyclohexanecarboxylic, cyclohexane-  
25

acetic, benzoic, methylbenzoic, t-butylbenzoic, methoxybenzoic, hydroxybenzoic, 3,5-di-t-butyl-4-hydroxybenzoic, phenylacetic, phenoxyacetic, 3-phenylpropionic, 3-(3,5-di-t-butyl-4-hydroxyphenyl)-propionic, furoic,  
5 tetrahydrofurancarboxylic, nicotinic, isonicotinic, pyroglutamic, 3-(2,2,6,6-tetramethyl-4-piperidylamino)crotonic, etc.

Examples of alkylene groups having not more than 12 carbon atoms are: methylene, ethylene, propylene, tri-  
10 methylene, tetramethylene, pentamethylene, 2,2-trimethyltrimethylene, hexamethylene, heptamethylene, octamethylene, trimethylhexamethylene, decamethylene, dodecamethylene, etc.

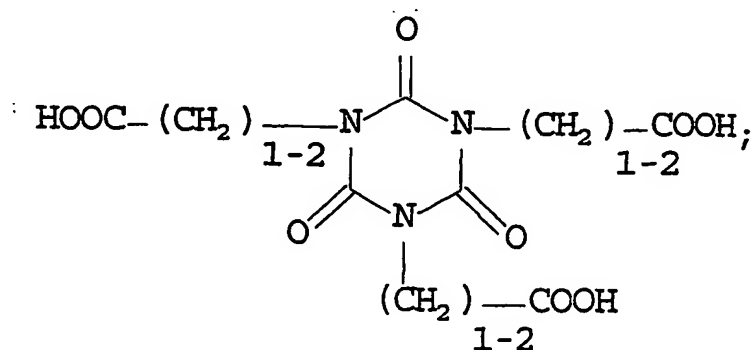
Examples of C<sub>4</sub>-C<sub>12</sub> alkylene groups interrupted by 1,  
15 2 or 3 oxygen atoms are: 3-oxapentane-1,5-diyl, 4-oxaheptane-1,7-diyl, 3,6-dioxa-octane-1,8-diyl, 4,7-dioxadecane-1,10-diyl, 4,9-dioxadodecane-1,12-diyl, 3,6,9-trioxaundecane-1,11-diyl, 4,7,10-trioxadecane-1,13-diyl, etc.

20 Examples of diacyl-aliphatic, cycloaliphatic, aromatic, arylaliphatic or heterocyclic groups having not more than 22 carbon atoms are those deriving from the following acids: oxalic, malonic, succinic, methylmalonic, allylmalonic, glutaric, ethylmalonic, adipic, pimelic,  
25 melic, diethylmalonic, suberic, azelaic, sebacic, 1,12-



dodecanoic, oxydiacetic, iminodiacetic, methyliminodiacetic, maleic, fumaric, itaconic, citraconic, cyclohexanedicarboxylic, cyclohexenedicarboxylic, bicycloheptenedicarboxylic, phthalic, isophthalic, terephthalic, phenylmalonic, benzylidenemalonic, benzylmalonic, butyl-3,5-dit-butyl-4-hydroxybenzylmalonic, furandicarboxylic, pyridinedicarboxylic, 1,4-dioxaspiro[4.5]decane-2,3-dicarboxylic, 1,5-dioxaspiro[5.5]undecane-3,3-dicarboxylic, 7,7,9,9-tetramethyl-1,4-dioxo-8-azaspiro[4.5]decane-2,3-dicarboxylic, 8,8,10,10-tetramethyl-1,5-dioxo-9-azaspiro[5.5]undecane-3,3-dicarboxylic, N-(2,2,6,6-tetramethyl-4-piperidyl)iminodiacetic, etc.

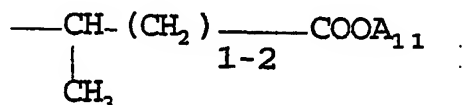
Examples of triacyl-aliphatic, aromatic or heterocyclic groups having not more than 18 carbon atoms are those deriving from the following acids: methanetricarboxylic, 1,1,2-ethanetricarboxylic, 1,2,3-propanetricarboxylic, 1,2,3-butanetricarboxylic, citric, nitrilotriacetic, benzenetricarboxylic, or from the acid having the formula:



etc.

Examples of tetra-acyl-aliphatic or aromatic groups having not more than 18 carbon atoms are those deriving from the following acids: 1,1,2,2-ethanetetracarboxylic, 1,1,3,3-propanetetracarboxylic, 1,2,2,3-propanetetracarboxylic, 1,2,3,4-butanetetracarboxylic, ethylenediaminetetra-acetic, pyromellitic, etc.

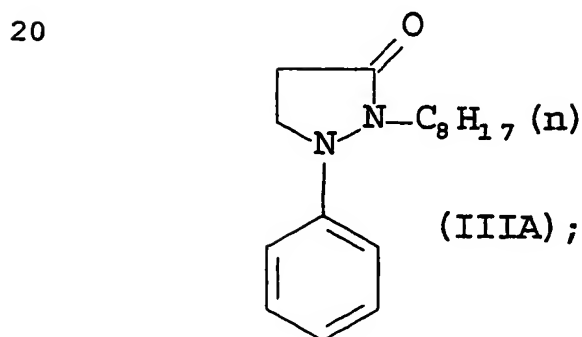
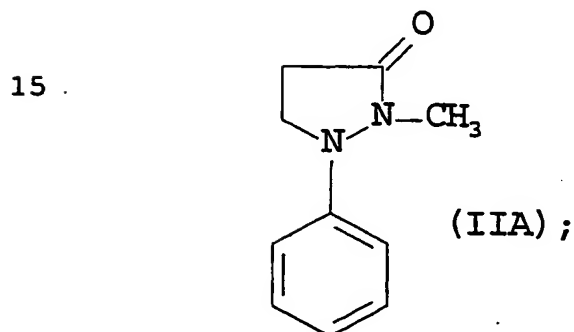
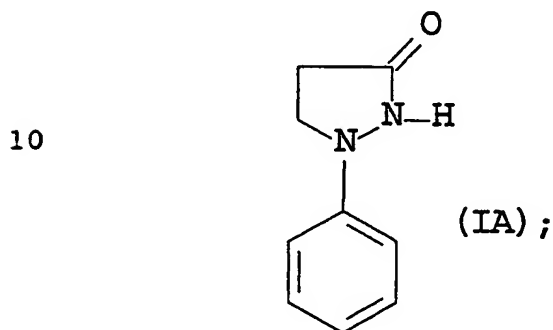
Compounds belonging to the group of 3-pyrazolidinones (a) having general formula (I) or (II), of interest for the purposes of the present invention, are those wherein  $A_1$  is a phenyl group, or a

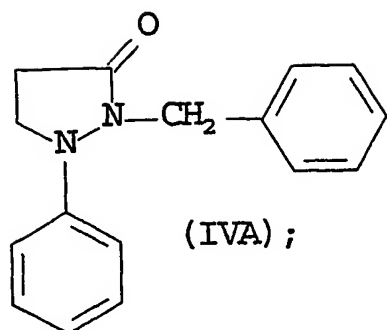


group, wherein  $A_{11}$  is a linear or branched  $C_{12}$ - $C_{18}$  alkyl group;  $A_3$ ,  $A_4$ ,  $A_5$  and  $A_6$  are hydrogen;  $s$  is 1 or 2; when  $s$  is 1,  $A_2$  is hydrogen, a linear or branched  $C_1$ - $C_{12}$  alkyl group, or a benzyl group; when  $s$  is 2,  $A_2$  is a xylylene group;  $A_7$  is a phenyl group;  $A_8$  is a hydrogen or a methyl;  $t$  is 1 or 2; when  $t$  is 1,  $A_9$  is a  $C_{10}$ - $C_{22}$  acyl-aliphatic group, a benzoyl group, or a group having general formula (IVa) or (IVb) wherein  $A_{13}$  is a linear or branched  $C_{12}$ - $C_{18}$  alkyl group,  $A_{14}$  is hydrogen,  $A_{15}$  is a linear or branched  $C_1$ - $C_4$  alkyl group or a cyclohexyl group; when  $t$  is 2,  $A_9$  is a  $-\text{CO}(\text{CH}_2)_{2-8}\text{CO}-$  group, or a group having general formula (Va) or (Vb) wherein  $A_{19}$  is

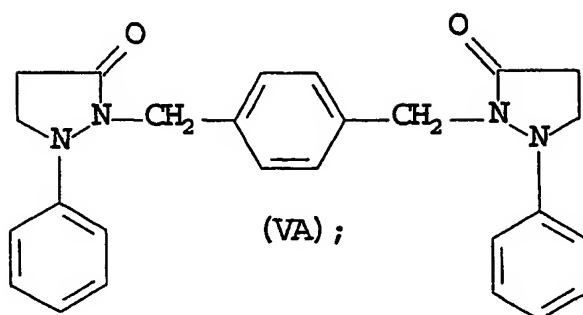
a linear or branched C<sub>4</sub>-C<sub>6</sub> alkylene group and A<sub>20</sub> has the same meanings defined above.

Compounds belonging to the group of 3-pyrazolidinones (a) having general formula (I) or (II) of particular interest for the purposes of the present invention but in no way limiting its scope, are:

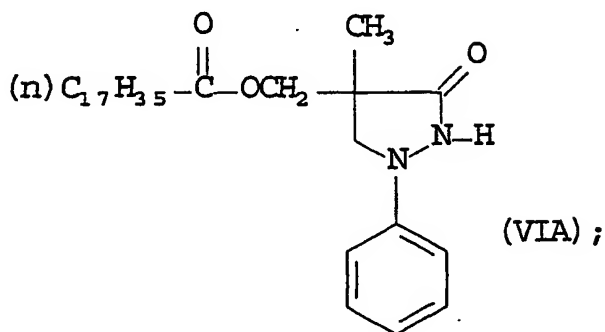




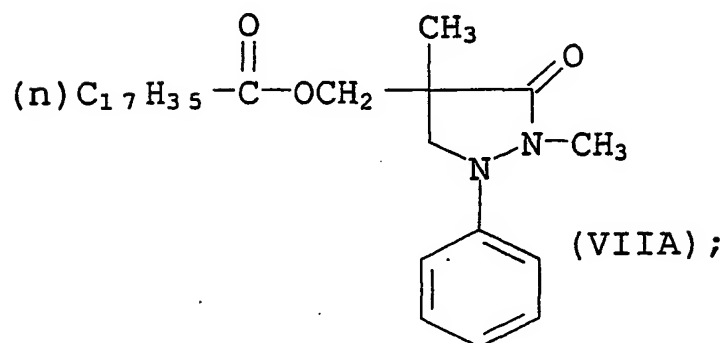
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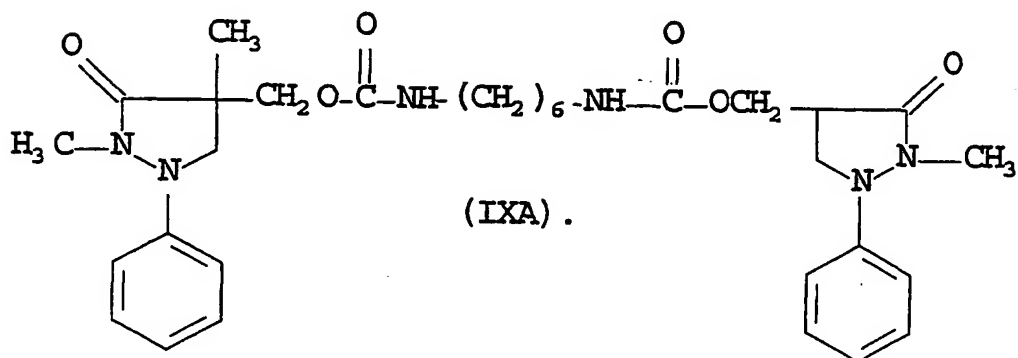
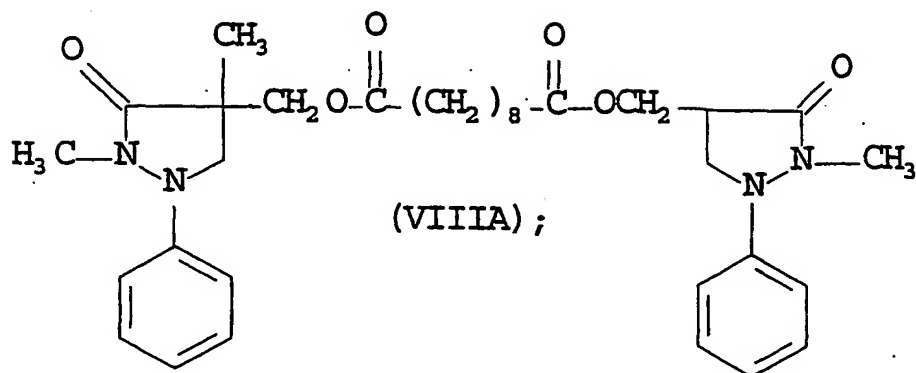


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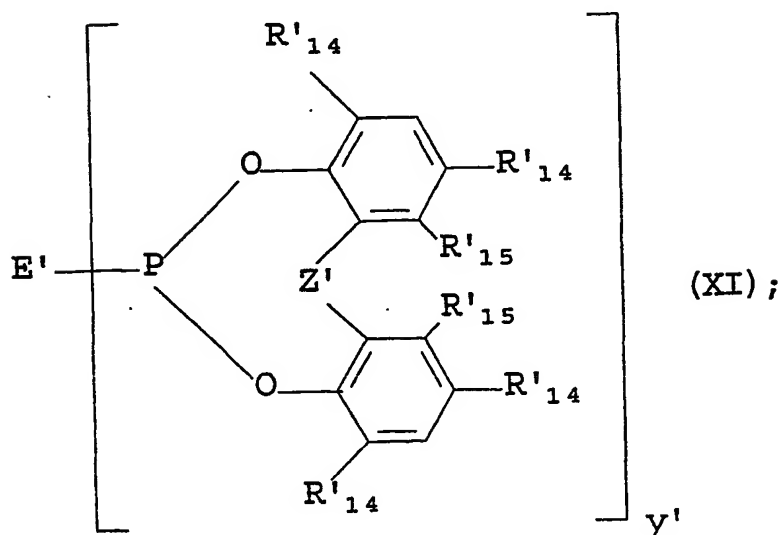
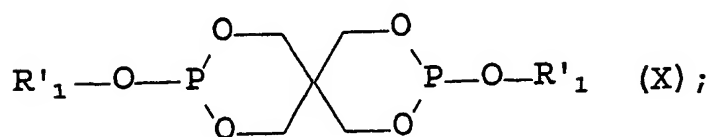
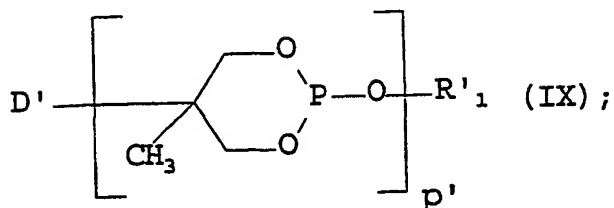
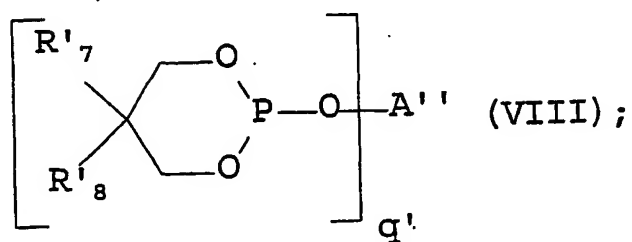
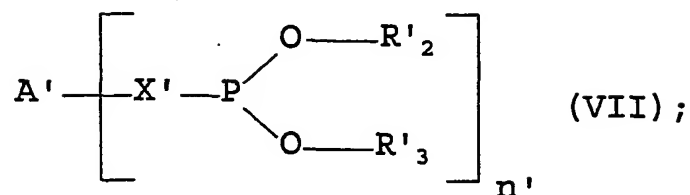
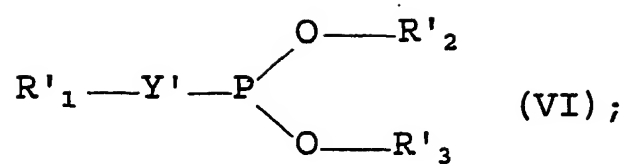
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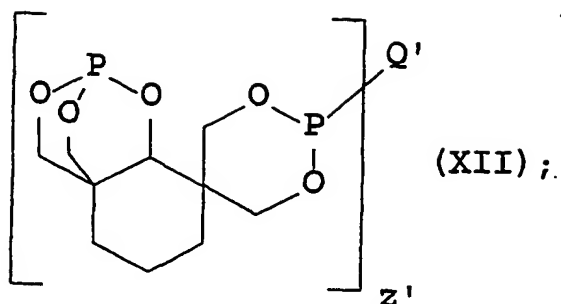
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The compounds belonging to the group of 3-pyrazolidinones (a) having general formula (I) or (II) are known in the art: some are commercially available products, others can be prepared as described, for example, in "Tetrahedron Letters" (1985), Vol. 26, page 5663; or in the following patents: DE 53,834, GB 650,911, JP 11/060559 and U.S. 4,835,285.

Compounds belonging to the group of organic phosphites or phosphonites (b) useful for the purposes of the present invention are selected from those having the following general formulae (VI)-(XII):



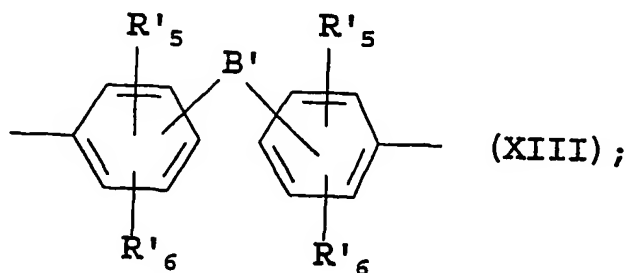


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wherein:

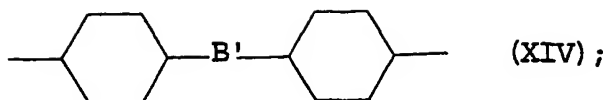
- $n'$  is 2, 3 or 4;
- $p'$  is 1 or 2;
- $q'$  is 2 or 3;
- 10 -  $r'$  is an integer ranging from 4 to 12, extremes included;
- $y'$  is 1, 2 or 3;
- $z'$  is an integer ranging from 1 to 6, extremes included;
- 15 - when  $n'$  is 2,  $A'$  represents a  $C_2-C_{18}$  alkylene group; a  $C_2-C_{12}$  alkylene group containing an oxygen atom, a sulfur atom, or an  $-NR'_4-$  group; a group having general formula (XIII):

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a group having general formula (XIV):

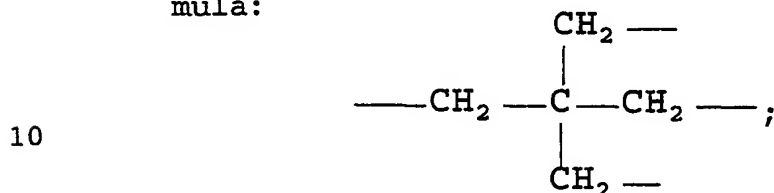
25



a phenylene group;

- when  $n'$  is 3,  $A'$  represents a group having general  
5 formula  $-C_{r'}H_{2r'-1}-$  wherein  $r'$  has the same values de-  
scribed above;

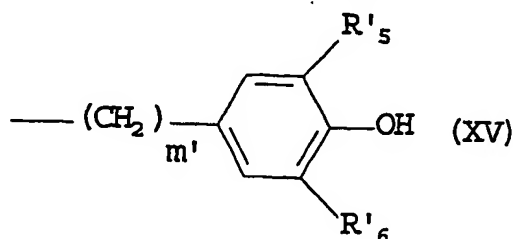
- when  $n'$  is 4,  $A'$  represents a group having the for-  
mula:



- $A''$  has the same meanings defined above for  $A'$  when  
 $n'$  is 2;
- $B'$  represents a direct bond; one of the following  
15 groups:  $-\text{CH}_2-$ ,  $-\text{CHR}'_4-$ ,  $-\text{CR}'_1\text{R}'_4-$ ; a sulfur atom; a  
 $\text{C}_5-\text{C}_7$  cycloalkylidene group; a cyclohexylidene group  
substituted with 1-4  $\text{C}_1-\text{C}_4$  alkyl groups in position  
3, 4 and/or 5;
- when  $p'$  is 1,  $D'$  represents a methyl group; and,  
20 when  $p'$  is 2,  $D'$  represents a  $-\text{CH}_2\text{OCH}_2-$  group;
- when  $y'$  is 1,  $E'$  represents a  $\text{C}_1-\text{C}_{18}$  alkyl group; an  
 $-\text{OR}'_1$  group; a halogen atom;
- when  $y'$  is 2,  $E'$  represents an  $-\text{O}-\text{A}''-\text{O}-$  group;
- when  $y'$  is 3,  $E'$  represents one of the following  
25 groups:  $\text{R}'_4\text{C}(\text{CH}_2\text{O}-)_3$ ,  $\text{N}(\text{CH}_2\text{CH}_2\text{O}-)_3$ ;



- Q' represents a radical of an alcohol or a phenol with valence z', said radical being attached to the phosphorous atom by means of an oxygen atom;
- R'<sub>1</sub>, R'<sub>2</sub> and R'<sub>3</sub>, each independently, represent a hydrogen atom; a halogen atom; one of the following groups: -COOR'<sub>4</sub>-, -CN-, -CONR'<sub>4</sub>R'<sub>4</sub>; a C<sub>2</sub>-C<sub>18</sub> alkyl group containing an oxygen atom, a sulfur atom, an -NR'<sub>4</sub>- group; a C<sub>7</sub>-C<sub>9</sub> phenylalkyl group; a C<sub>5</sub>-C<sub>12</sub> cycloalkyl group; a phenyl group or a naphthyl group, said phenyl or naphthyl groups optionally substituted with halogen atoms, or with 1-3 C<sub>1</sub>-C<sub>18</sub> alkyl or alkoxy groups or with C<sub>7</sub>-C<sub>9</sub> phenylalkyl groups; or they represent a group having general formula (XV):



wherein m' represents an integer ranging from 3 to 6, extremes included;

- 20 - R'<sub>4</sub> represents a hydrogen atom; a C<sub>1</sub>-C<sub>18</sub> alkyl group; a C<sub>5</sub>-C<sub>12</sub> cycloalkyl group; a C<sub>7</sub>-C<sub>9</sub> phenylalkyl group;
- R'<sub>5</sub> and R'<sub>6</sub>, each independently, represent a hydrogen atom; a C<sub>1</sub>-C<sub>8</sub> alkyl group; a C<sub>5</sub>-C<sub>6</sub> cycloalkyl group;
- 25 - when q' is 2, R'<sub>7</sub> and R'<sub>8</sub>, each independently, represent a C<sub>1</sub>-C<sub>4</sub> alkyl group; or, considered jointly,

- they represent a 2,3-dihdropentamethylene group;
- when  $q'$  is 3,  $R'_7$  and  $R'_8$  represent a methyl group;
  - $R'_{14}$  represents a hydrogen atom; a  $C_1-C_9$  alkyl group; a cyclohexyl group;
  - 5 -  $R'_{15}$  represents a hydrogen atom; a methyl group; or, when two or more  $R'_{14}$  and  $R'_{15}$  groups are present, said groups are the same as or different from each other;
  - $X'$  and  $Y'$  represent a direct bond; an oxygen atom;
  - 10 -  $Z'$  represents a direct bond; a methylene group; a  $-C(R'_{16})_2-$  group; a sulfur atom;
  - $R'_{16}$  represents a  $C_1-C_8$  alkyl group.

Compounds belonging to the group of organic phosphites or phosphonites (b) of particular interest for

15 the purposes of the present invention are those having general formula (VI), (VII), (X) and (XI) wherein:

- $n'$  is 2 and  $y'$  is 1, 2 or 3;
- $A'$  represents a  $C_2-C_{18}$  alkylene group; a p-phenylene or p-bisphenylene group;
- 20 - when  $y'$  is 1,  $E'$  represents a  $C_1-C_{18}$  alkyl group; an  $-OR'-$  group, a fluorine atom;
- when  $y'$  is 2,  $E'$  represents a p-bisphenylene group;
- when  $y'$  is 3,  $E'$  represents an  $N(CH_2CH_2O-)_3$  group;
- $R'_1$ ,  $R'_2$  and  $R'_3$ , each independently represent a
- 25  $C_1-C_{18}$  alkyl group; a  $C_7-C_9$  phenylalkyl group; a cy-

clohexyl group; a phenyl group; said phenyl group optionally substituted with 1-3 C<sub>1</sub>-C<sub>18</sub> alkyl groups;

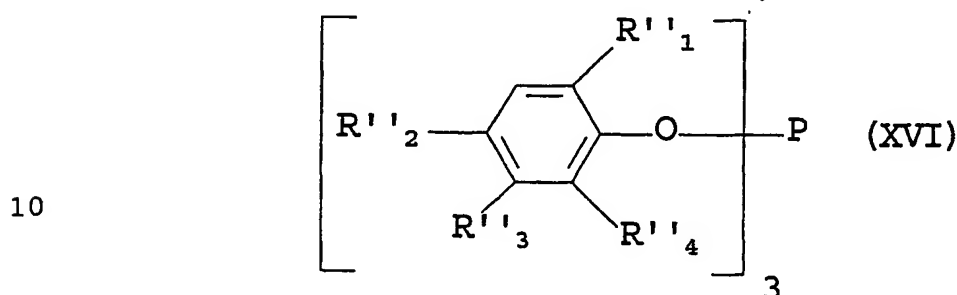
- R'<sub>14</sub> represents a hydrogen atom; a C<sub>1</sub>-C<sub>9</sub> alkyl group;
- R'<sub>15</sub> represents a hydrogen atom; a methyl group;
- 5 - X' represents a direct bond;
- Y' represents an oxygen atom;
- Z' represents a direct bond; a -CH(R'<sub>16</sub>)- group;
- R'<sub>16</sub> represents a C<sub>1</sub>-C<sub>4</sub> alkyl group.

Similarly, compounds belonging to the group of organic phosphites or phosphonites (b) of particular interest for the purposes of the present invention are those having general formula (VI), (VII), (X) and (XI) wherein:

- n' is 2 and y' is 1 or 3;
- A' represents a p-bisphenylene group;
- 15 - when y' is 1, E' represents a C<sub>1</sub>-C<sub>18</sub> alkoxyl group; a fluorine atom;
- when y' is 3, E' represents an N(CH<sub>2</sub>CH<sub>2</sub>O-)<sub>3</sub> group;
- R'<sub>1</sub>, R'<sub>2</sub> and R'<sub>3</sub>, each independently, represent a C<sub>1</sub>-C<sub>18</sub> alkyl group; a phenyl group substituted with
- 20 2-3 C<sub>2</sub>-C<sub>12</sub> alkyl groups;
- R'<sub>14</sub> represents a methyl group; a t-butyl group;
- R'<sub>15</sub> represents a hydrogen atom;
- X' represents a direct bond;
- Y' represents an oxygen atom;
- 25 - Z' represents a direct bond; a methylene group; a

-CH(CH<sub>3</sub>)- group.

Preferred compounds belonging to the group of organic phosphites or phosphonites (b) for the purposes of the present invention are those having general formula (VI), (VII) and (XI); particularly preferred are those  
 5 (VI), (VII) and (XI); particularly preferred are those having general formula (XVI):



wherein:

- R'<sub>1</sub> and R'<sub>2</sub>, each independently, represent a hydrogen atom; a C<sub>1</sub>-C<sub>8</sub> alkyl group; a cyclohexyl  
 15 group; a phenyl group;
- R'<sub>3</sub> and R'<sub>4</sub>, each independently, represent a hydrogen atom; a C<sub>1</sub>-C<sub>4</sub> alkyl group.

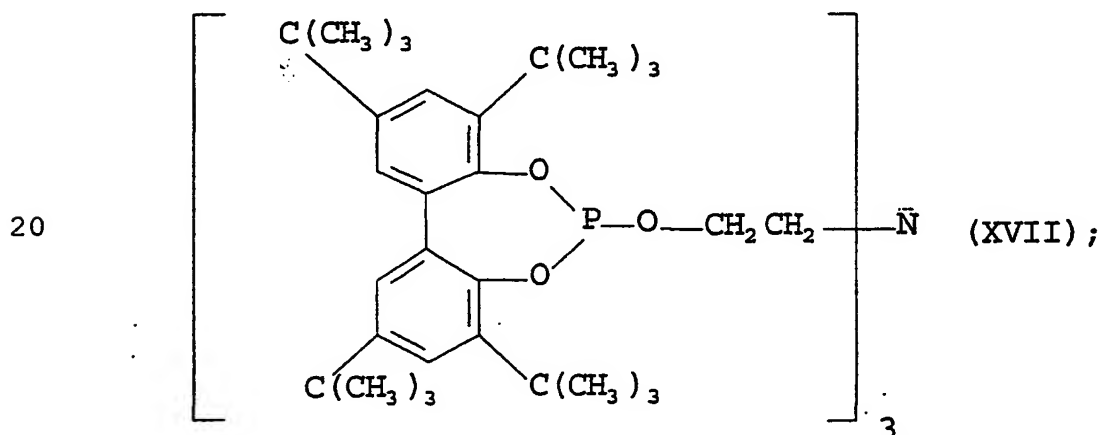
Specific examples of compounds belonging to the group of phosphites or phosphonites (b) useful for the purposes of the present invention are: triphenyl phosphite; diphenyl alkyl phosphites; phenyl dialkyl phosphites; tris(nonylphenyl)phosphite (known under the trade-name of Alkanox<sup>TM</sup> TNPP of Great Lakes Chemical Corporation); trilauryl phosphite; trioctadecyl phosphite;  
 20 distearyl pentaerythritol diphosphite; tris(2,4-di-t-

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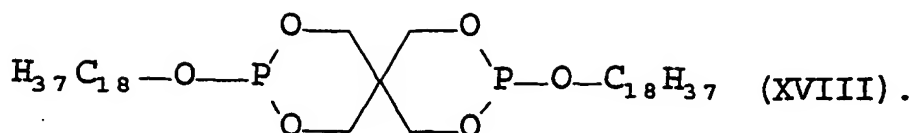
butylphenyl)phosphite (known under the trade-name of Alkanox™ 240 of Great Lakes Chemical Corporation); diisodecyl pentaerythritol diphosphite; bis(2,4-di-t-butylphenyl)pentaerythritol diphosphite (known under the trade-name of Alkanox™ P24 of Great Lakes Chemical Corporation); bis(2,6-di-t-butyl-4-methylphenyl)pentaerythritol diphosphite; bis-isodecyloxypentaerythritol diphosphite, bis(2,4-di-t-butyl-6-methylphenyl)pentaerythritol diphosphite; bis(2,4,6-tris-t-butylphenyl)pentaerythritol diphosphite; tristearylsorbitol triphosphite; tetra-kis(2,4-di-t-butylphenyl)-4,4'-diphenylenediphosphonite (known under the trade-name of Alkanox™ 24-44 of Great Lakes Chemical Corporation); 6-iso-octyloxy-2,4,8,10-tetra-t-butyl-12H-dibenzo-[d,g]-1,3,2-dioxaphosphocine; 6-fluoro-2,4,8,10-tetra-t-butyl-12-methyldibenzo[d,g]-1,3,2-dioxaphosphocine; bis(2,4-di-t-butyl-6-methylphenyl)methyl phosphite; bis-(2,4-di-t-butyl-6-methylphenyl)ethyl phosphite (known under the trade-name of Irgafos® 38 of Ciba Specialty Chemicals); bis(2,4-dicumylphenyl)pentaerythritol diphosphite.

Specific examples of particularly preferred compounds belonging to the group of organic phosphites or phosphonites (b) for the purposes of the present invention are: tris(2,4-di-t-butylphenyl)phosphite (known under the trade-name of Alkanox™ 240 of Great Lakes Chemi-

cal Corporation); tris(nonylphenyl)phosphite (known under the trade-name of Alkanox<sup>TM</sup> TNPP of Great Lakes Chemical Corporation); 6-fluoro-2,4,8,10-tetra-t-butyl-12-methyl-dibenzo[d,g]-1,3,2-dioxaphosphocine; 6-iso-octyloxy-2,4,-  
 5 8,10-tetra-t-butyl-12H-dibenzo-[d,g]-1,3,2-dioxaphosphocine; bis(2,4-di-t-butylphenyl)pentaerythritol diphosphite (known under the trade-name of Alkanox<sup>TM</sup> P24 of Great Lakes Chemical Corporation); bis(2,6-di-t-butyl-4-methylphenyl)pentaerythritol diphosphite; bis-(2,4-di-t-  
 10 butyl-6-methylphenyl) ethyl phosphite (known under the trade-name of Irgafos<sup>®</sup> 38 of Ciba Specialty Chemicals); bis-(2,4-dicumylphenyl) pentaerythritol diphosphite; tetrakis(2,4-di-t-butylphenyl)-4,4'-diphenylenediphosphonite (known under the trade-name of Alkanox<sup>TM</sup> 24-44 of  
 15 Great Lakes Chemical Corporation); triphosphite having formula (XVII):



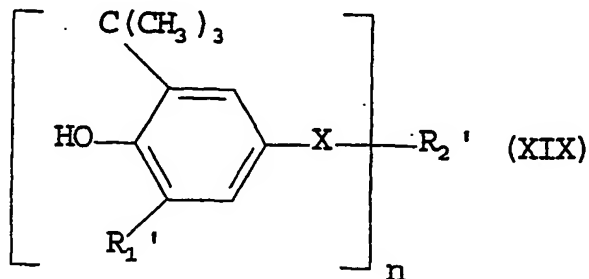
diphosphite having general formula (XVIII):



Specific examples of compounds belonging to the group of organic phosphites or phosphonites (b) even more preferred for the purposes of the present invention are: tris(2,4-di-t-butylphenyl)phosphite (known under the trade-name of Alkanox™ 240 of Great-Lakes Chemical Corporation); bis-(2,4-di-t-butyl-6-methylphenyl)ethyl phosphite (known under the trade-name of Irgafos® 38 of Ciba Specialty Chemicals); tetrakis(2,4-di-t-butylphenyl)-4,4'-diphenylenediphosphonite (known under the trade-name of Alkanox™ 24-44 of Great Lakes Chemical Corporation); bis(2,4-dicumylphenyl)pentaerythritol diphosphite.

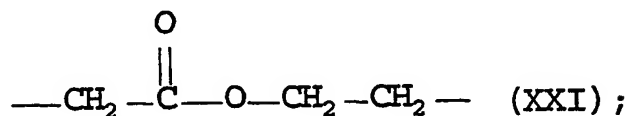
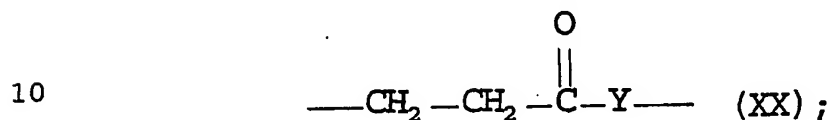
The compounds belonging to the group of organic phosphites or phosphonites (b) described above are known compounds; many of them are commercially available products.

Compounds belonging to the group of sterically hindered phenols (c) useful for the purposes of the present invention are selected from those having general formula (XIX):



wherein:

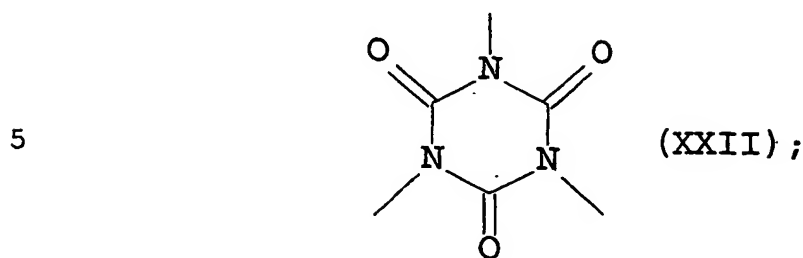
- 5 -  $R_1'$  represents a  $C_1$ - $C_4$  alkyl group;
- $n$  is 1, 2, 3 or 4;
- $X$  represents a methylene group; or a group having general formula (XX) or (XXI):



- $Y$  represents an oxygen atom; an  $\text{-NH-}$  group;
- 15 - when  $n$  is 1,  $X$  represents a group having general formula (XX) wherein  $Y$  is attached to  $R_2'$  and  $R_2'$  represents a  $C_1$ - $C_{25}$  alkyl group;
- when  $n$  is 2,  $X$  represents a group having general formula (XX) wherein  $Y$  is attached to  $R_2'$  and  $R_2'$  represents a  $C_2$ - $C_{12}$  alkylene group; a  $C_4$ - $C_{12}$  alkylene group containing one or more oxygen or sulfur atoms; or, when  $Y$  represents an  $\text{-NH-}$  group,  $R_2'$  represents a direct bond;
- 20 - when  $n$  is 3,  $X$  represents a methylene group; a group having general formula (XXI) wherein the ethylene
- 25



group is bound to  $R_2'$  and  $R_2'$  represents a group having formula (XXII):



- when  $n$  is 4,  $X$  represents a group having general formula (XX) wherein  $Y$  is bound to  $R_2'$  and  $R_2'$  represents a  $C_4$ - $C_{10}$  alkane-tetrayl group.

Examples of linear or branched alkyl groups having up to 25 carbon atoms are: methyl, ethyl, propyl, isopropyl,  $n$ -butyl,  $s$ -butyl, isobutyl,  $t$ -butyl, 2-ethylbutyl,  $n$ -pentyl, isopentyl, 1-methylpentyl, 1,3-dimethylbutyl,  $n$ -hexyl, 1-methylhexyl,  $n$ -heptyl, isoheptyl, 1,1,3,3-tetramethylbutyl, 1-methylheptyl, 3-methylheptyl,  $n$ -octyl, 2-ethylhexyl, 1,1,3-trimethylhexyl, 1,1,3,3-tetramethylpentyl, nonyl, decyl, undecyl, 1-methylundecyl, dodecyl, 1,1,3,3,5,5-hexamethylhexyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, eicosyl or doicosyl, etc.

$R_1'$  preferably represents methyl or  $t$ -butyl.

$R_2'$  preferably represents a  $C_1$ - $C_{20}$  alkyl group, particularly a  $C_1$ - $C_{18}$  alkyl group, for example, a  $C_4$ - $C_{18}$  alkyl group. Even more preferably,  $R_2'$  represents a  $C_8$ - $C_{18}$  alkyl

group, particularly a C<sub>14</sub>-C<sub>18</sub> alkyl group, for example octadecyl.

Examples of linear or branched C<sub>2</sub>-C<sub>12</sub> alkylene groups, are: ethylene, propylene, tetramethylene, pentamethylene, hexamethylene, heptamethylene, octamethylene, decamethylene, dodecamethylene, etc.

R<sub>2</sub>' preferably represents a C<sub>2</sub>-C<sub>10</sub> alkylene group, particularly a C<sub>2</sub>-C<sub>8</sub> alkylene group. Even more preferably, R<sub>2</sub>' represents a C<sub>4</sub>-C<sub>8</sub> alkylene group, particularly a C<sub>4</sub>-C<sub>6</sub> alkylene group, for example hexamethylene.

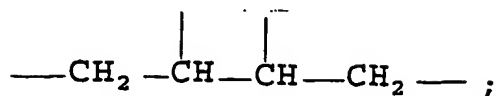
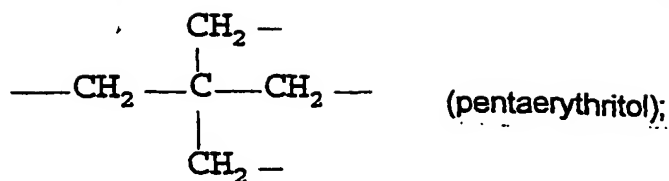
Examples of C<sub>4</sub>-C<sub>12</sub> alkylene groups containing one or more oxygen or sulfur atoms, are:

-CH<sub>2</sub>-O-CH<sub>2</sub>CH<sub>2</sub>-O-CH<sub>2</sub>-, -CH<sub>2</sub>-(O-CH<sub>2</sub>CH<sub>2</sub>)<sub>2</sub>-O-CH<sub>2</sub>-,  
-CH<sub>2</sub>-(O-CH<sub>2</sub>CH<sub>2</sub>)<sub>3</sub>-O-CH<sub>2</sub>-, -CH<sub>2</sub>-(O-CH<sub>2</sub>CH<sub>2</sub>)<sub>4</sub>-O-CH<sub>2</sub>-,  
-CH<sub>2</sub>-CH<sub>2</sub>-O-CH<sub>2</sub>CH<sub>2</sub>-O-CH<sub>2</sub>CH<sub>2</sub>-, -CH<sub>2</sub>-CH<sub>2</sub>-S-CH<sub>2</sub>CH<sub>2</sub>-, etc.

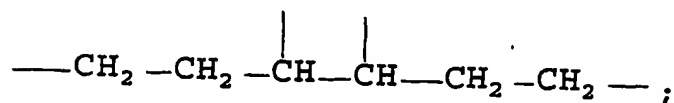
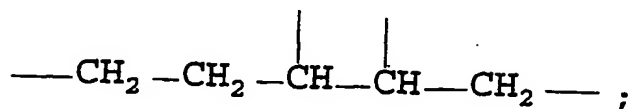
R<sub>2</sub>' preferably represents a C<sub>4</sub>-C<sub>10</sub> alkylene group containing one or more oxygen or sulfur atoms, particularly a C<sub>4</sub>-C<sub>8</sub> alkylene group containing one or more oxygen or sulfur atoms, for example a C<sub>4</sub>-C<sub>6</sub> alkylene group containing one or more oxygen or sulfur atoms. Even more preferably, R<sub>2</sub>' represents one of the following groups:

-CH<sub>2</sub>-CH<sub>2</sub>-O-CH<sub>2</sub>CH<sub>2</sub>-O-CH<sub>2</sub>CH<sub>2</sub>-, -CH<sub>2</sub>-CH<sub>2</sub>-S-CH<sub>2</sub>CH<sub>2</sub>-.

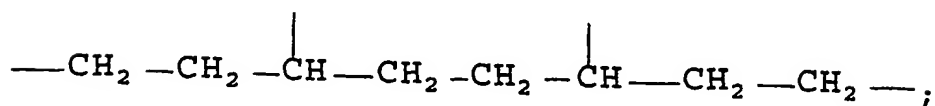
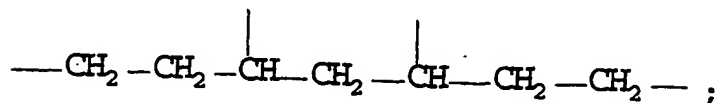
Examples of C<sub>4</sub>-C<sub>10</sub> alkane-tetrayl groups are:



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etc. Pentaerythritol is preferred.

Specific examples of compounds belonging to the  
 15 group of sterically hindered phenols (c) useful for the  
 purposes of the present invention are those wherein, in  
 general formula (XIX), when  $n$  is 1,  $R_2'$  represents a  $C_1$ -  
 $C_{20}$  alkyl group.

Specific examples of preferred compounds belonging  
 20 to the group of sterically hindered phenols (c) for the  
 purposes of the present invention are those wherein, in  
 general formula (XIX), when  $n$  is 2,  $R_2'$  represents a  $C_2$ - $C_8$   
 alkylene group, or a  $C_4$ - $C_8$  alkylene group containing one  
 or more oxygen or sulfur atoms; or, when  $Y$  represents an  
 25 -NH- group,  $R_2'$  represents a direct bond; and, when  $n$  is

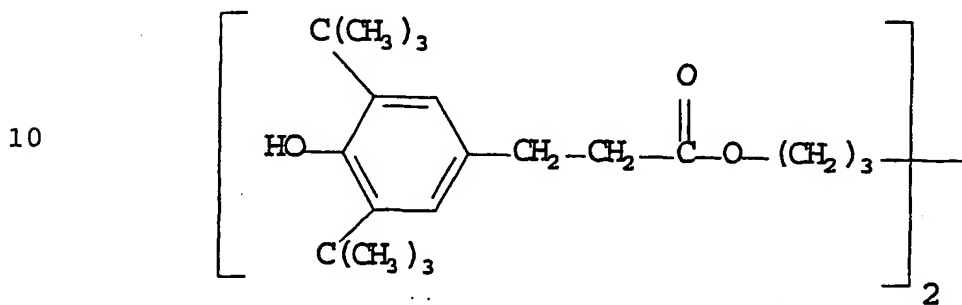
4, R<sub>2</sub>' represents a C<sub>4</sub>-C<sub>8</sub> alkane-tetrayl group.

Similarly, examples of preferred compounds belonging to the group of sterically hindered phenols (c) for the purposes of the present invention are those wherein, in  
5 general formula (XIX), R<sub>1</sub>' represents methyl or t-butyl; n is 1, 2 or 4; X represents a group having general formula (XX); Y represents an oxygen atom or an -NH- group; and, when n is 1, R<sub>2</sub>' represents a C<sub>14</sub>-C<sub>18</sub> alkyl group; and, when n is 2, R<sub>2</sub>' represents a C<sub>4</sub>-C<sub>6</sub> alkylene group,  
10 or a C<sub>4</sub>-C<sub>6</sub> alkylene group containing one or more oxygen or sulfur atoms; and, when n is 4, R<sub>2</sub>' represents a C<sub>4</sub>-C<sub>6</sub> alkane-tetrayl group.

Similarly, of interest for the purposes of the present invention are compounds belonging to the group of  
15 sterically hindered phenols (c) having general formula (XIX), selected from: N,N'-hexamethylene bis[3-(3,5-di-t-butyl-4-hydroxyphenyl)propionamide] (known under the trade-name of Lowinox<sup>®</sup> HD-98 of Great Lakes Chemical Corporation); octadecyl 3-(3',5'-di-t-butyl-4'-hydroxyphenyl)propionate (known under the trade-name of Anox<sup>™</sup> PP18  
20 of Great Lakes Chemical Corporation); tetrakis(methylene-(3,5-di-t-butyl-4-hydroxy-hydrocinnamate)methane (known under the trade-name of Anox<sup>™</sup> 20 of Great Lakes Chemical Corporation); triethylene glycol bis[3-(3-t-butyl-4-hydroxy-5-methylphenyl)propionate] (known under the  
25

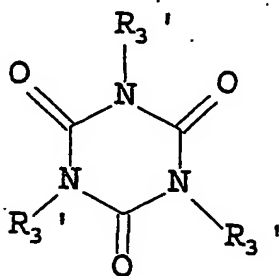
trade-name of Lowinox® GP-45 of Great Lakes Chemical Corporation); 2,2'-thiodiethylene bis[3(3,5-di-t-butyl-4-hydroxyphenyl)propionate] (known under the trade-name of Anox™ 70 of Great Lakes Chemical Corporation); 1,3,5-

5 tris(3,5-di-t-butyl-4-hydroxybenzyl)isocyanurate (known under the trade-name of Anox™ IC-14 of Great Lakes Chemical Corporation); the compound having the formula:

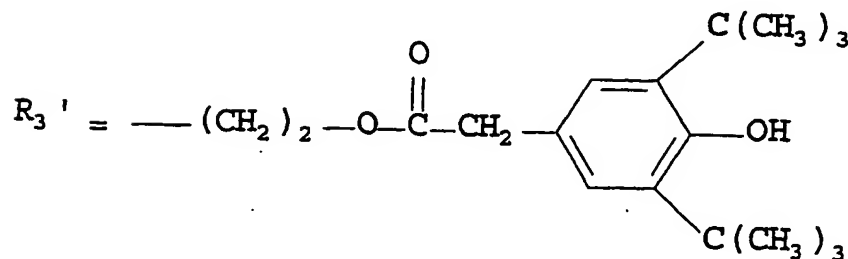


(known under the trade-name of Irganox® 259 of Ciba Specialty Chemicals); the compound having the formula:

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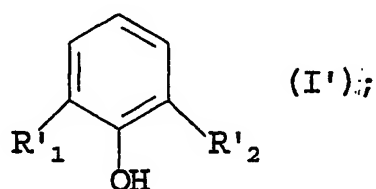
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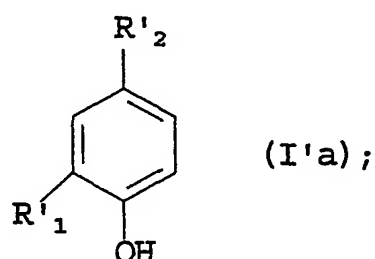
(known under the trade-name of Irganox® 3125 of Ciba Specialty Chemicals).

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Also of interest for the purposes of the present invention are compounds belonging to the group of sterically hindered phenols (c) consisting of reactive antioxidant compounds containing a sterically hindered phenol group having general formula (I') or (I'a):

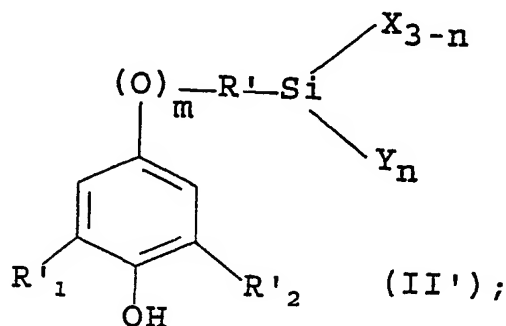


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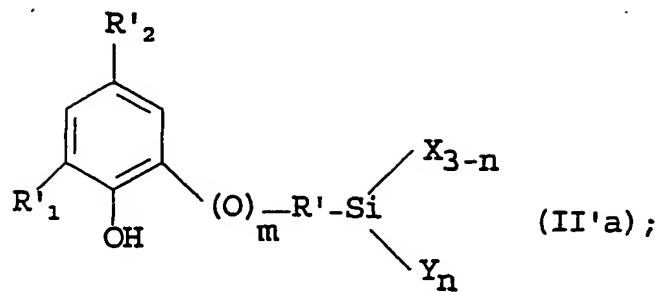


more specifically, reactive antioxidant compounds containing sterically hindered phenol groups having general formula (I') or (I'a) useful for the purposes of the present invention, are selected from those having general formula (II') or (II'a):

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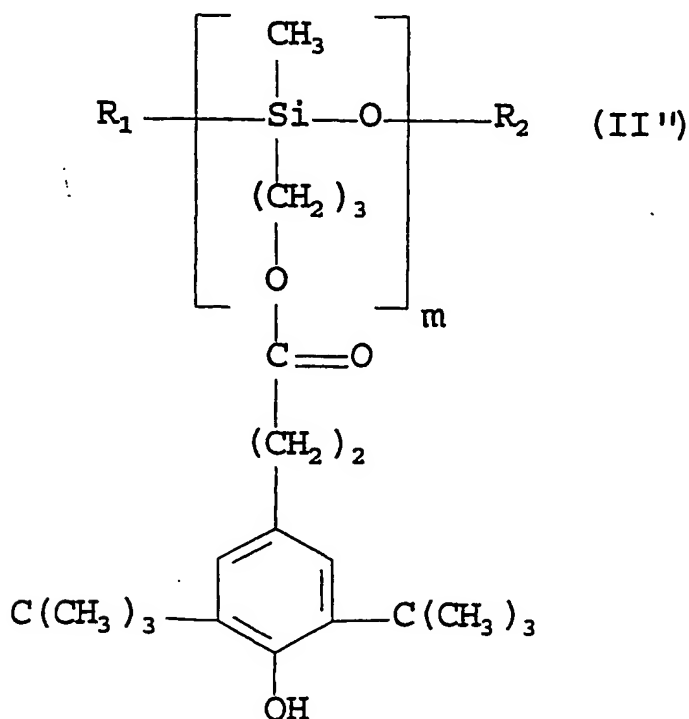


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said reactive antioxidant compounds containing sterically  
 5 hindered phenol groups having general formula (I'),  
 (I'a), (II') and (II'a), are described in European pat-  
 ents EP 162,523 and EP 182,415 which should be considered  
 as forming an integrant part of the present patent appli-  
 cation. The above reactive antioxidant compounds may pro-  
 10 duce complex resinous structures by means of hydrolysis  
 and condensation of the hydrolyzable silicic function.

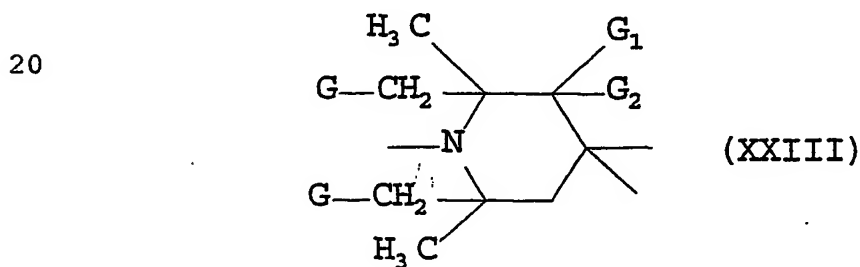
A specific example of the above reactive anti-  
 oxidant compounds is the following compound having gen-  
 eral formula (II''):



consisting of a mixture of linear oligomers wherein  $R_1 =$   
5 OH and  $R_2 = H$ , and cyclic oligomers wherein  $R_1$  and  $R_2$  represent a direct bond, having a number average molecular weight equal to 3,900.

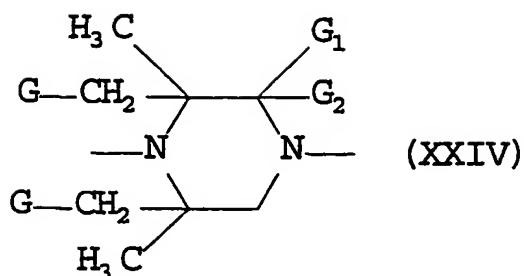
As said above, the compounds belonging to the sterically hindered phenols (c) group are known compounds and,  
10 in some cases, are commercially available. Or, said compounds, can be prepared according to processes described, for example, in patents U.S. 3,330,859, U.S. 3,960,928, or in European patents EP 162,523 and EP 182,415.

Compounds belonging to the group of sterically hindered amines (d) useful for the purposes of the present  
15 invention are selected from those comprising at least one group having general formula (XXIII) or (XXIV):



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wherein:

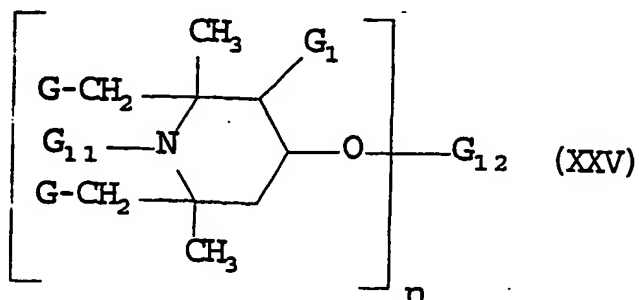
- G represents a hydrogen atom; or a methyl group;
- G<sub>1</sub> and G<sub>2</sub>, the same or different, represent a hydrogen atom; a methyl group; or, they jointly represent an oxygen atom.

10

Of particular interest for the purposes of the present invention are compounds belonging to the group of sterically hindered amines (d) selected from compounds (a')-(h') which comprise at least one group having general formula (XXIII) or (XXIV).

15

(a') Compounds having general formula (XXV):



20

wherein n is a number ranging from 1 to 4, extremes included; G and G<sub>1</sub>, independently represent a hydrogen atom

25

or a methyl;  $G_{11}$  represents a hydrogen atom, O, a hydroxyl group, an NO group, a  $-\text{CH}_2\text{CN}$  group, a  $\text{C}_1\text{-C}_{18}$  alkyl group, a  $\text{C}_3\text{-C}_8$  alkenyl group, a  $\text{C}_3\text{-C}_8$  alkynyl group, a  $\text{C}_7\text{-C}_{12}$  arylalkyl group, a  $\text{C}_1\text{-C}_{18}$  alkoxyl group, a  $\text{C}_5\text{-C}_8$  cycloalkoxyl group, a  $\text{C}_7\text{-C}_9$  phenylalkoxyl group, a  $\text{C}_1\text{-C}_8$  alkanoyl group, a  $\text{C}_3\text{-C}_5$  alkenoyl group, a  $\text{C}_1\text{-C}_{18}$  alkanoyloxyl group, a benzyloxyl group, a glycidyl group, an  $\text{OG}_{11}'$  group, wherein  $G_{11}'$  represents a linear or branched  $\text{C}_1\text{-C}_{10}$  alkyl group; a  $-\text{CH}_2\text{CH}(\text{OH})-\text{Z}$  group wherein Z represents a hydrogen atom, a methyl, or a phenyl,  $G_{11}$  preferably being hydrogen, a  $\text{C}_1\text{-C}_4$  alkyl group, an allyl, a benzyl, an acetyl or an acryloyl;  $G_{12}$ , when n is 1, represents a hydrogen atom, a  $\text{C}_1\text{-C}_{18}$  alkyl group, said alkyl group optionally containing one or more oxygen atoms, a cyanoethyl group, a benzyl, a glycidyl group, a monovalent radical of a carboxylic acid, of a carbamic acid or of an acid containing phosphorous, aliphatic, cycloaliphatic or arylaliphatic, unsaturated or aromatic, or a monovalent silyl radical, preferably a radical of an aliphatic carboxylic acid having from 2 to 18 carbon atoms, of a cycloaliphatic carboxylic acid having from 7 to 15 carbon atoms, of an  $\alpha,\beta$ -unsaturated carboxylic acid having from 3 to 5 carbon atoms, of an aromatic carboxylic acid having from 7 to 15 carbon atoms, said carboxylic acids optionally substituted in the aliphatic, cycloaliphatic or

aromatic part, with 1-3  $-\text{COOZ}_{12}$  groups wherein  $Z_{12}$  represents a hydrogen atom, a  $\text{C}_1\text{-C}_{20}$  alkyl group, a  $\text{C}_3\text{-C}_{12}$  alkenyl group, a  $\text{C}_5\text{-C}_7$  cycloalkyl group, a phenyl or a benzyl;  $G_{12}$ , when  $n$  is 2, represents a  $\text{C}_2\text{-C}_{12}$  alkylene group, a  $\text{C}_4\text{-C}_{12}$  alkenylene group, a xylylene group, a divalent radical of a dicarboxylic acid, of a dicarbamic acid or of an acid containing phosphorous, aliphatic, cycloaliphatic, arylaliphatic or aromatic, or a divalent silyl radical, preferably a radical of an aliphatic dicarboxylic acid having from 2 to 36 carbon atoms, of a cycloaliphatic or aromatic dicarboxylic acid having from 8 to 14 carbon atoms, of an aliphatic, cycloaliphatic or aromatic dicarbamic acid, having from 8 to 14 carbon atoms, said dicarboxylic acids optionally substituted in the aliphatic, cycloaliphatic or aromatic part, with 1 or 2  $-\text{COOZ}_{12}$  groups wherein  $Z_{12}$  has the same meanings defined above;  $G_{12}$ , when  $n$  is 3, represents a trivalent radical of an aliphatic, cycloaliphatic or aromatic tricarboxylic acid, optionally substituted in the aliphatic, cycloaliphatic or aromatic part, with a  $-\text{COOZ}_{12}$  group wherein  $Z_{12}$  has the same meanings defined above, or of an aromatic tricarbamic acid or of an acid containing phosphorous, or it represents a trivalent silyl radical;  $G_{12}$ , when  $n$  is 4, is a tetravalent radical of an aliphatic, cycloaliphatic or aromatic tetracarboxylic acid.

Radicals of tetracarboxylic acids comprise, in all cases, radicals having the formula  $(-CO)_nR$  wherein  $n$  has the same meaning described above and  $R$  can be easily deduced from the above definition.

5        Examples of  $C_1$ - $C_{12}$  alkyl groups are: methyl, ethyl, n-propyl, n-butyl, s-butyl, t-butyl, n-hexyl, n-octyl, 2-ethylhexyl, n-nonyl, n-decyl, n-undecyl, n-dodecyl, etc.

      Examples of  $G_{11}$  and  $G_{12}$  substituents, when they represent a  $C_1$ - $C_{18}$  alkyl group, are: in addition to the  
10       groups described above: n-tridecyl, n-tetradecyl, n-hexadecyl, n-octadecyl, etc.

      Examples of the  $G_{11}$  substituent, when it represents a  $C_1$ - $C_8$  alkenyl group, are: 1-propenyl, allyl, methallyl, 2-butenyl, 2-pentenyl, 2-hexenyl, 2-octenyl, 4-t-butyl-2-  
15       butenyl, etc.

      When the  $G_{11}$  substituent represents a  $C_3$ - $C_8$  alkynyl group, it is preferably propargyl.

      When the  $G_{11}$  substituent represents a  $C_7$ - $C_{12}$  arylalkyl group, it is, in particular, phenethyl, preferably  
20       benzyl.

      Examples of the  $G_{11}$  substituent, when it represents a  $C_1$ - $C_8$  alkanoyl group, are: formyl, propionyl, butyryl, octanoyl, preferably acetyl and, when it represents a  $C_3$ - $C_5$  alkenoyl group, preferably acryloyl.

25       Examples of the  $G_{12}$  substituent, when it represents

a monovalent radical of a carboxylic acid, are radicals of the following acids: acetic, caproic, stearic, acrylic, methacrylic, benzoic,  $\beta$ -(3,5-di-t-butyl-4-hydroxyphenyl)propionic, etc.

5        An example of the  $G_{12}$  substituent, when it represents a monovalent silyl radical, is: a radical having general formula  $-(C_jH_{2j})-Si(Z')_jZ''$  wherein  $j$  is an integer ranging from 2 to 5, extremes included, and  $Z'$  and  $Z''$ , each independently, represent a  $C_1$ - $C_4$  alkyl group or  
10    a  $C_1$ - $C_4$  alkoxyl group.

Examples of the  $G_{12}$  substituent, when it represents a divalent radical of a dicarboxylic acid, are radicals of the following acids: malonic, succinic, glutaric, adipic, suberic, sebacic, maleic, itaconic, phthalic, di-  
15    butylmalonic, dibenzylmalonic, butyl(3,5-di-t-butyl-4-hydroxybenzyl)malonic, bicycloheptenedicarboxylic, etc.

Examples of the  $G_{12}$  substituent, when it represents a trivalent radical of a tricarboxylic acid, are radicals of the acids: trimellitic, citric, nitrilotriacetic, etc.

20        Examples of the  $G_{12}$  substituent, when it represents a tetravalent radical of a tetracarboxylic acid, are radicals of the following acids: butane-1,2,3,4-tetracarboxylic, pyromellitic, etc.

Examples of the  $G_{12}$  substituent, when it represents  
25    a divalent radical of a dicarbamic acid, are radicals of

the following acids: hexamethylenedicarbamic, 2,4-toluylenedicarbamic, etc.

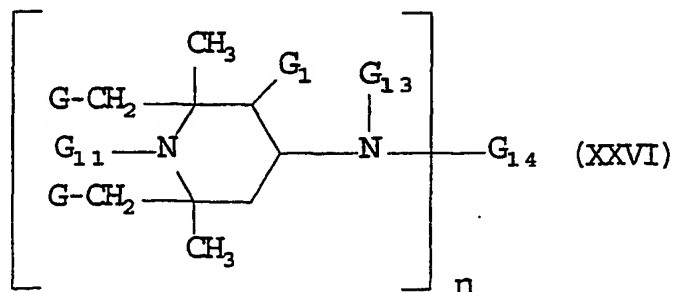
Compounds having general formula (XXV) wherein G represents a hydrogen atom,  $G_{11}$  represents a hydrogen atom or a methyl, n is 2 and  $G_{12}$  represents a diacyl radical of an aliphatic dicarboxylic acid having from 4 to 12 carbon atoms, are preferred.

Specific examples of polyalkylpiperidines having general formula (XXV) are:

- 10 1) 4-hydroxy-2,2,6,6-tetramethylpiperidine;
- 2) 1-allyl-4-hydroxy-2,2,6,6-tetramethylpiperidine;
- 3) 1-benzyl-4-hydroxy-2,2,6,6-tetramethylpiperidine;
- 4) 1-(4-t-butyl-2-butenyl)-4-hydroxy-2,2,6,6-tetramethylpiperidine;
- 15 5) 4-stearyloxy-2,2,6,6-tetramethylpiperidine;
- 6) 1-ethyl-4-salicyloyloxy-2,2,6,6-tetramethylpiperidine;
- 7) 4-methacryloyloxy-1,2,2,6,6-pentamethylpiperidine;
- 8) 1,2,2,6,6-pentamethylpiperidin-4-yl- $\beta$ -(3,5-di-t-butyl-4-hydroxyphenyl)propionate;
- 20 9) di(1-benzyl-2,2,6,6-tetramethylpiperidin-4-yl)maleate;
- 10) di(2,2,6,6-tetramethylpiperidin-4-yl)succinate;
- 11) di(2,2,6,6-tetramethylpiperidin-4-yl)glutarate;
- 25 12) di(2,2,6,6-tetramethylpiperidin-4-yl)adipate;

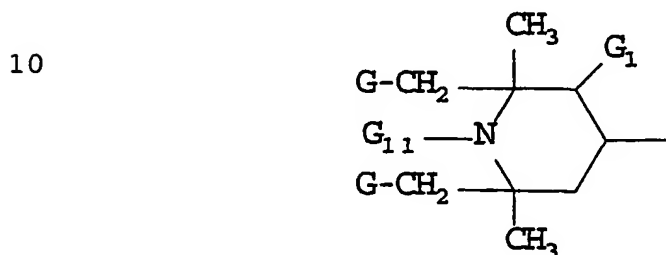
- 13) di (2,2,6,6-tetramethylpiperidin-4-yl) sebacate;
- 14) di (1,2,2,6,6-pentamethylpiperidin-4-yl) sebacate;
- 15) di (1,2,3,6-tetramethyl-2,6-diethylpiperidin-4-yl) -  
sebacate;
- 5 16) di (1-allyl-2,2,6,6-tetramethylpiperidin-4-yl) phthal-  
ate;
- 17) 1-hydroxy-4- $\beta$ -cyanoethyloxy-2,2,6,6-tetramethylpip-  
eridine;
- 18) (1-acetyl-2,2,6,6-tetramethylpiperidin-4-yl) acetate;
- 10 19) tris (2,2,6,6-tetramethylpiperidin-4-yl) trimellitate;
- 20) 1-acryloyl-4-benzyloxy-2,2,6,6-tetramethylpiperid-  
ine;
- 21) di (2,2,6,6-tetramethylpiperidin-4-yl) diethylmalon-  
ate;
- 15 22) di (1,2,2,6,6-pentamethylpiperidin-4-yl) dibutylmalon-  
ate;
- 23) di (1,2,2,6,6-pentamethylpiperidin-4-yl) butyl- (3,5-  
di-tert-butyl-4-hydroxybenzyl) malonate;
- 24) di (1-octyloxy-2,2,6,6-tetramethylpiperidin-4-yl) seb-  
20 acate;
- 25) di (1-cyclohexyloxy-2,2,6,6-tetramethylpiperidin-4-  
yl) sebacate;
- 26) hexane-1',6'-bis (4-carbamoyloxy-1-n-butyl-2,2,6,6-  
tetramethylpiperidine);
- 25 27) toluene-2',4'-bis (4-carbamoyloxy-1-n-propyl-2,2,6,6-

- tetramethylpiperidine);
- 28) dimethyl-bis(2,2,6,6-tetramethylpiperidin-4-oxy)silane;
- 29) phenyl-tris(2,2,6,6-tetramethylpiperidin-4-oxy)silane;
- 30) tris(1-propyl-2,2,6,6-tetramethylpiperidin-4-yl)-phosphite;
- 31) tris(1-propyl-2,2,6,6-tetramethylpiperidin-4-yl)-phosphate;
- 32) bis(1,2,2,6,6-pentamethylpiperidin-4-yl)phenylphosphonate;
- 33) 4-hydroxy-1,2,2,6,6-pentamethylpiperidine;
- 34) 4-hydroxy-N-hydroxyethyl-2,2,6,6-tetramethylpiperidine;
- 35) 4-hydroxy-N-(2-hydroxypropyl)-2,2,6,6-tetramethylpiperidine;
- 36) 1-glycidyl-4-hydroxy-2,2,6,6-tetramethylpiperidine.
- (b') Compounds having general formula (XXVI):





wherein n is 1 or 2; G, G<sub>1</sub> and G<sub>11</sub> have the same meanings described above under point (a'); G<sub>13</sub> represents a hydrogen atom, a C<sub>1</sub>-C<sub>12</sub> alkyl group, a C<sub>2</sub>-C<sub>5</sub> hydroxyalkyl group, a C<sub>5</sub>-C<sub>7</sub> cycloalkyl group, a C<sub>7</sub>-C<sub>8</sub> arylalkyl group, a C<sub>2</sub>-C<sub>18</sub> alkanoyl group, a C<sub>3</sub>-C<sub>5</sub> alkenoyl group, a benzoyl group, or a group having the following general formula:



wherein G, G<sub>1</sub> and G<sub>11</sub> have the same meanings defined above under point (a'); G<sub>14</sub>, when n is 1, represents a hydrogen atom, a C<sub>1</sub>-C<sub>18</sub> alkyl group, a C<sub>3</sub>-C<sub>8</sub> alkenyl group, a C<sub>5</sub>-C<sub>7</sub> cycloalkyl group, a C<sub>1</sub>-C<sub>4</sub> alkyl group substituted with a hydroxyl group, with a cyano group, with an alkoxy carbonyl group or with a carbamide group, a glycidyl group, a group having the formula -CH<sub>2</sub>-CH(OH)-Z or a group having the formula -CONH-Z wherein Z represents a hydrogen atom, a methyl or a phenyl; G<sub>14</sub>, when n is 2, represents a C<sub>2</sub>-C<sub>12</sub> alkylene group, a C<sub>6</sub>-C<sub>12</sub> arylene group, a xylylene group, a group having the formula -CH<sub>2</sub>-CH(OH)-CH<sub>2</sub>- or a group having the formula -CH<sub>2</sub>-CH(OH)-CH<sub>2</sub>-O-D-O wherein D

represents a C<sub>2</sub>-C<sub>10</sub> alkylene group, a C<sub>6</sub>-C<sub>15</sub> arylene group,  
a C<sub>6</sub>-C<sub>12</sub> cycloalkylene group; or, on the condition that  
G<sub>13</sub> does not represent an alkanoyl group, an alkenoyl  
group or a benzoyl group, G<sub>14</sub> can also represent a 1-oxo-  
5 (C<sub>2</sub>-C<sub>12</sub>)-alkylene group, a divalent radical of a dicarbox-  
ylic acid or of a dicarbamic acid, aliphatic, cyclo-  
aliphatic or aromatic, or also a -CO- group; or, when n  
is 1, G<sub>13</sub> and G<sub>14</sub> considered jointly, can also represent a  
divalent radical of a 1,2- or 1,3-dicarboxylic acid, ali-  
10 phatic, cycloaliphatic or aromatic.

The C<sub>1</sub>-C<sub>12</sub> alkyl or C<sub>1</sub>-C<sub>18</sub> alkyl groups have already  
been described above under point (a').

The C<sub>5</sub>-C<sub>7</sub> cycloalkyl groups are preferably cyclo-  
hexyl.

15 When G<sub>13</sub> represents a C<sub>7</sub>-C<sub>8</sub> arylalkyl group, it is  
phenylethyl, preferably benzyl.

When G<sub>13</sub> represents a C<sub>2</sub>-C<sub>5</sub> hydroxyalkyl group, it is  
2-hydroxyethyl, 2-hydroxypropyl, etc.

Examples of G<sub>13</sub>, when it represents a C<sub>2</sub>-C<sub>18</sub> alkanoyl  
20 group, are: propionyl, butyryl, octanoyl, dodecanoyl,  
hexadecanoyl, octadecanoyl, etc. preferably acetyl and,  
when it represents a C<sub>3</sub>-C<sub>5</sub> alkenoyl group, it is prefera-  
bly acryloyl.

Examples of G<sub>14</sub>, when it represents a C<sub>2</sub>-C<sub>8</sub> alkenyl  
25 group, are: allyl, methallyl, 2-butenyl, 2-pentenyl, 2-

hexenyl, 2-octenyl, etc.

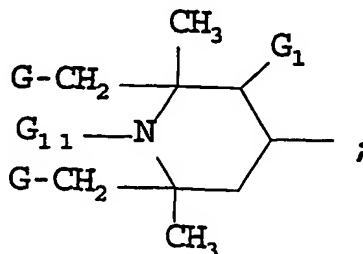
Examples of  $G_{14}$ , when it represents a  $C_1$ - $C_4$  alkyl group substituted by a hydroxyl, cyano, alkoxycarbonyl or carbamide group, are: 2-hydroxyethyl, 2-hydroxypropyl, 2-cyanoethyl, methoxycarbonylmethyl, 2-ethoxycarbonylethyl, 2-aminocarbonylpropyl, 2-(dimethylaminocarbonyl)ethyl, etc.

Examples of  $C_2$ - $C_{12}$  alkylene groups, are: ethylene, propylene, 2,2-dimethylpropylene, tetramethylene, hexamethylene, octamethylene, decamethylene, dodecamethylene, etc.

Examples of  $C_6$ - $C_{15}$  arylene groups, are: o-, m- or p-phenylene, 1,4-naphthylene, 4,4'-diphenylene, etc.

An example of a  $C_6$ - $C_{12}$  cycloalkylene group is preferably cyclohexylene.

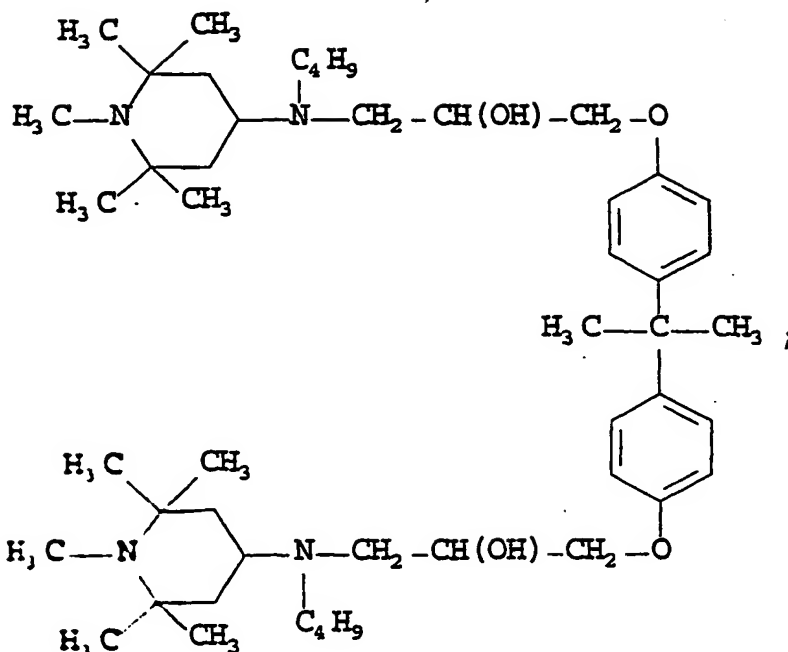
Preferred compounds having general formula (XXVI) are those wherein n is 1 or 2, G represents a hydrogen atom,  $G_{11}$  represents a hydrogen atom or a methyl,  $G_{13}$  represents a hydrogen atom, a  $C_1$ - $C_{12}$  alkyl group or a group having the formula:



and G<sub>14</sub>, when n is 1, represents a hydrogen atom or a C<sub>1</sub>-C<sub>12</sub> alkyl group and, when n is 2, it represents a C<sub>2</sub>-C<sub>8</sub> alkylene group or a 1-oxo-(C<sub>2</sub>-C<sub>8</sub>)-alkylene group.

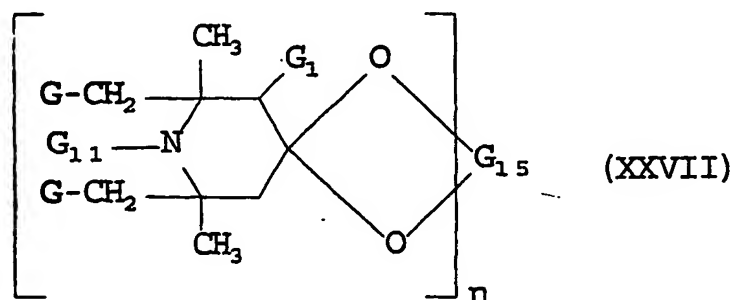
5        Specific examples of polyalkylpiperidines having general formula (XXVI) are:

- 1) N,N'-bis(2,2,6,6-tetramethylpiperidin-4-yl)hexamethylene-1,6-diamine;
- 2) N,N'-bis(2,2,6,6-tetramethylpiperidin-4-yl)hexamethylene-1,6-diacetamide;
- 10        3) bis(2,2,6,6-tetramethylpiperidin-4-yl) amine;
- 4) 4-benzoylamino-2,2,6,6-tetramethylpiperidine;
- 5) N,N'-bis(2,2,6,6-tetramethylpiperidin-4-yl)-N,N'-di-butyladipamide;
- 15        6) N,N'-bis(2,2,6,6-tetramethylpiperidin-4-yl)-N,N'-di-cyclohexyl-2-hydroxypropylene-1,3-diamine;
- 7) N,N'-bis(2,2,6,6-tetramethylpiperidin-4-yl)-p-xylylenediamine;
- 8) N,N'-bis(2,2,6,6-tetramethylpiperidin-4-yl)succinamide;
- 20        9) bis(2,2,6,6-tetramethylpiperidin-4-yl)-N-(2,2,6,6-tetramethylpiperidin-4-yl)- $\beta$ -aminodipropionate;
- 10) compound having the formula:



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- 10 11) 4-[bis(2-hydroxyethyl)amino]-1,2,2,6,6-pentamethyl-  
piperidine;
- 12) 4-(3-methyl-4-hydroxy-5-tert-butylbenzamide)-2,2,-  
6,6-tetramethylpiperidine;
- 13) 4-methacrylamide-1,2,2,6,6-pentamethylpiperidine;
- 15 (c') Compounds having general formula (XXVII):



20

wherein n is 1 or 2; G, G<sub>1</sub> and G<sub>11</sub> have the same meanings described above under point (a'); G<sub>15</sub>, when n is 1, represents a C<sub>2</sub>-C<sub>8</sub> alkylene or hydroxyalkylene group, or a C<sub>4</sub>-C<sub>22</sub> acyloxyalkylene group, and, when n is 2, it is a

25 (-CH<sub>2</sub>)<sub>2</sub>C(CH<sub>2</sub>-)<sub>2</sub> group.

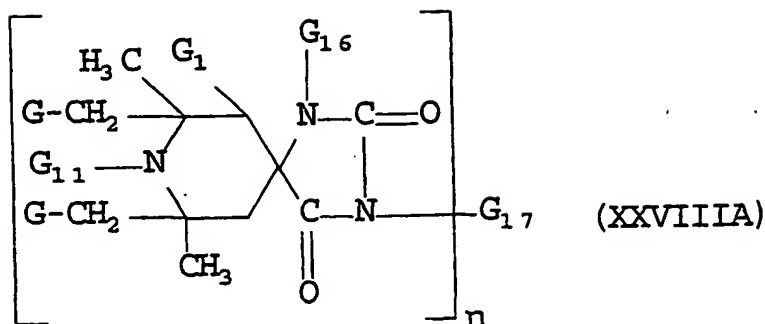
Examples of  $G_{15}$ , when it represents a  $C_2-C_8$  alkylene or hydroxyalkylene group, are: ethylene, 1-methylethylene, propylene, 2-ethylpropylene, 2-ethyl-2-hydroxymethyl-propylene, etc.

5        An example of  $G_{15}$ , when it represents a  $C_4-C_{22}$  acyloxyalkylene group is 2-ethyl-2-acetoxy-methylpropylene.

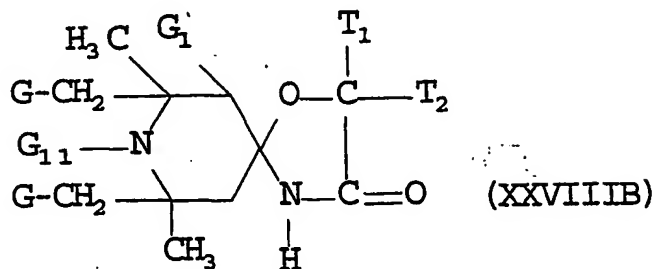
Specific examples of polyalkylpiperidines having general formula (XXVII) are:

- 1) 9-aza-8,8,10,10-tetramethyl-1,5-dioxaspiro[5.5]un-  
10        decane;
- 2) 9-aza-8,8,10,10-tetramethyl-3-ethyl-1,5-dioxaspiro-  
      [5.5]undecane;
- 3) 8-aza-2,7,7,8,9,9-hexamethyl-1,4-dioxaspiro[4.5]-  
      decane;
- 15    4) 9-aza-3-hydroxymethyl-3-ethyl-8,8,9,10,10-penta-  
      methyl-1,5-dioxaspiro-[5.5]undecane;
- 5) 9-aza-3-ethyl-3-acetoxymethyl-9-acetyl-8,8,10,10-  
      tetramethyl-1,5-dioxaspiro[5.5]undecane;
- 6) 2,2,6,6-tetramethylpiperidine-4-spiro-2'-(1',3'-  
20        dioxane)-5'-spiro-5'''-(1''',3'''-dioxane)-2'''-spiro-  
      4'''-(2''',2''',6''',6'''-tetramethylpiperidine).

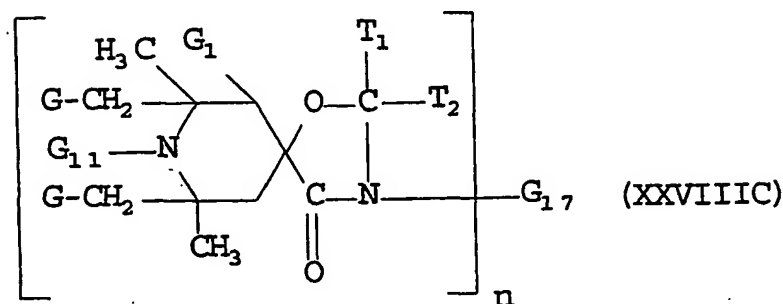
(d') Compounds having general formula (XXVIII A), (XXVIII B) and (XXVIII C), compounds having general formula (XXVIII C) being preferred:



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- 15 wherein  $n$  is 1 or 2,  $G$ ,  $G_1$  and  $G_{11}$  have the same meanings defined above under point (a');  $G_{16}$  represents a hydrogen atom, a  $C_1$ - $C_{12}$  alkyl group, an allyl group, a benzyl, a glycidyl group or a  $C_2$ - $C_6$  alkoxyalkyl group;  $G_{17}$ , when  $n$  is 1, represents a hydrogen atom, a  $C_1$ - $C_{12}$  alkyl group, a  $C_3$ - $C_5$  alkenyl group, a  $C_7$ - $C_9$  arylalkyl group, a  $C_5$ - $C_7$  cycloalkyl group, a  $C_2$ - $C_4$  hydroxyalkyl group, a  $C_2$ - $C_6$  alkoxyl group, a  $C_6$ - $C_{10}$  aryl group, a glycidyl group, or a group having the formula  $-(\text{CH}_2)_p\text{-COO-Q}$  or  $-(\text{CH}_2)_p\text{-O-CO-Q}$  wherein  $p$  is 1 or 2 and  $Q$  represents a  $C_1$ - $C_4$  alkyl group
- 20 or a phenyl;  $G_{17}$ , when  $n$  is 2, represents a  $C_2$ - $C_{12}$  al-
- 25

kylene group, a C<sub>4</sub>-C<sub>12</sub> alkenylene group, a C<sub>6</sub>-C<sub>12</sub> arylene group, a group having the formula:

-CH<sub>2</sub>-CH(OH)-CH<sub>2</sub>-O-D-O-CH<sub>2</sub>-CH(OH)-CH<sub>2</sub>- wherein D represents a C<sub>1</sub>-C<sub>10</sub> alkylene group, a C<sub>6</sub>-C<sub>15</sub> arylene group, a C<sub>6</sub>-C<sub>12</sub>

5 cycloalkylene group, or a group having the formula:

-CH<sub>2</sub>-CH(OZ')-CH<sub>2</sub>-(OCH<sub>2</sub>-CH(OZ')-CH<sub>2</sub>)<sub>2</sub> wherein Z' represents a hydrogen atom, a C<sub>1</sub>-C<sub>18</sub> alkyl group, an allyl, a benzyl, or a C<sub>1</sub>-C<sub>4</sub> alkanoyl group or a benzoyl; T<sub>1</sub> and T<sub>2</sub> each

independently represent a hydrogen atom, a C<sub>1</sub>-C<sub>18</sub> alkyl group, a C<sub>4</sub>-C<sub>10</sub> aryl group, a C<sub>7</sub>-C<sub>9</sub> arylalkyl group, said groups optionally substituted with a halogen atom or with a C<sub>1</sub>-C<sub>4</sub> alkyl group; or T<sub>1</sub> and T<sub>2</sub>, considered jointly with the carbon atom to which they are bound, form a C<sub>5</sub>-C<sub>14</sub> cycloalkane ring.

15 Examples of C<sub>1</sub>-C<sub>12</sub> alkyl groups are: methyl, ethyl, n-propyl, n-butyl, s-butyl, t-butyl, n-hexyl, n-octyl, 2-ethylhexyl, n-nonyl, n-decyl, n-undecyl, n-dodecyl, etc.

Examples of C<sub>1</sub>-C<sub>18</sub> alkyl groups are, in addition to those listed above: n-tridecyl, n-tetradecyl, n-hexadecyl, n-octadecyl; etc.

Examples of C<sub>2</sub>-C<sub>6</sub> alkoxyalkyl groups are: methoxymethyl, ethoxymethyl, propoxymethyl, t-butoxymethyl, ethoxyethyl, ethoxypropyl, n-butoxyethyl, t-butoxyethyl, isopropoxyethyl, propoxypropyl, etc.

25 Examples of G<sub>17</sub>, when it represents a C<sub>3</sub>-C<sub>5</sub> alkenyl



group are: 1-propenyl, allyl, methallyl, 2-butenyl, 2-pentenyl, etc.

Examples of  $G_{17}$ ,  $T_1$  and  $T_2$ , when they represent a  $C_7$ - $C_9$  arylalkyl group are: phenethyl, preferably benzyl.

5        Examples of cycloalkane rings, formed by  $T_1$  and  $T_2$  when said substituents are considered jointly with the carbon atom to which they are bound, are: cyclopentane, cyclohexane, cyclo-octane, cyclododecane, etc.

10       Examples of  $G_{17}$ , when it represents a  $C_2$ - $C_4$  hydroxyalkyl group, are: 2-hydroxyethyl, 2-hydroxypropyl, 2-hydroxybutyl, 4-hydroxybutyl, etc.

15       Examples of  $G_{17}$ ,  $T_1$  and  $T_2$ , when they represent a  $C_6$ - $C_{10}$  aryl group are: phenyl,  $\alpha$ - or  $\beta$ -naphthyl, optionally substituted with a halogen atom or a  $C_1$ - $C_4$  alkyl group, etc.

Examples of  $G_{17}$ , when it represents a  $C_2$ - $C_{12}$  alkylene group, are: ethylene, propylene, 2,2-dimethylpropylene, tetramethylene, hexamethylene, octamethylene, decamethylene, dodecamethylene, etc.

20       Examples of  $G_{17}$ , when it represents a  $C_4$ - $C_{12}$  alkenylene group, are: 2-butenylene, 2-pentenylene, 3-hexenylene, etc.

25       Examples of  $G_{17}$ , when it represents a  $C_6$ - $C_{12}$  arylene group, are: o-, m- or p-phenylene, 1,4-naphthylene, 4,4'-diphenylene, etc.

Examples of Z', when it represents a C<sub>2</sub>-C<sub>12</sub> alkanoyl group, are: propionyl, butyryl, octanoyl, dodecanoyl, preferably acetyl.

Examples of D, when it represents a C<sub>2</sub>-C<sub>10</sub> alkylene group, a C<sub>6</sub>-C<sub>15</sub> arylene group or C<sub>6</sub>-C<sub>12</sub> cycloalkylene group, are described above relating to point (b').

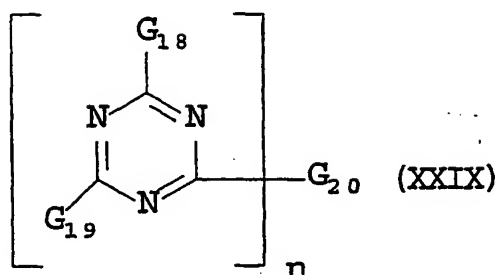
Specific examples of polyalkylpiperidines having general formula (XXVIII A), (XXVIII B) and (XXVIII C) are:

- 1) 3-benzyl-1,3,8-triaza-7,7,9,9-tetramethylspiro[4.5]-  
10 decane-2,4-dione;
- 2) 3-n-octyl-1,3,8-triaza-7,7,9,9-tetramethylspiro-  
[4.5]-decane-2,4-dione;
- 3) 3-allyl-1,3,8-triaza-1,7,7,9,9-pentamethylspiro-  
[4.5]-decane-2,4-dione;
- 15 4) 3-glycidyl-1,3,8-triaza-7,7,8,9,9-pentamethylspiro-  
[4.5]-decane-2,4-dione;
- 5) 1,3,7,7,8,9,9-heptamethyl-1,3,8-triazaspiro[4.5]-  
decane-2,4-dione;
- 6) 2-isopropyl-7,7,9,9-tetramethyl-1-oxa-3,8-diaza-4-  
20 oxospiro[4.5]decane;
- 7) 2,2-dibutyl-7,7,9,9-tetramethyl-1-oxa-3,8-diaza-4-  
oxospiro[4.5]decane;
- 8) 2,2,4,4-tetramethyl-7-oxa-3,20-diaza-21-oxodispiro-  
[5.1.11.2]heneicosane;
- 25 9) 2-butyl-7,7,9,9-tetramethyl-1-oxa-4,8-diaza-3-oxo-



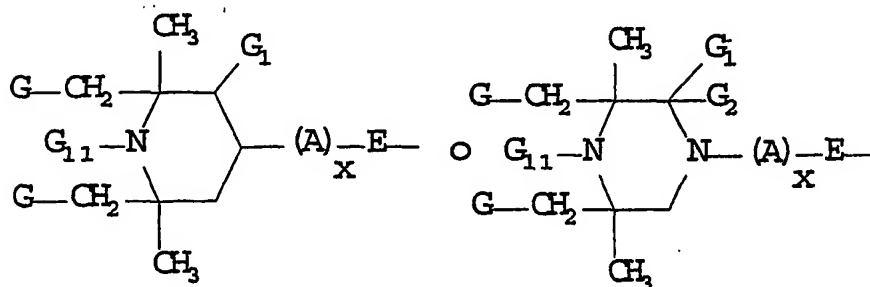
(e') Compounds having general formula (XXIX):

5



10 wherein n is 1 or 2, and G<sub>18</sub> represents a group having one of the following formulae:

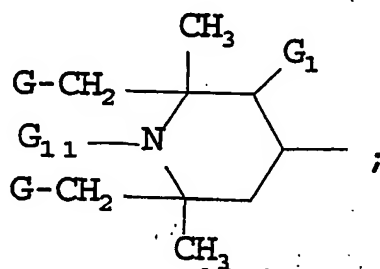
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wherein G and G<sub>11</sub> have the same meanings defined above under point (a'); G<sub>1</sub> and G<sub>2</sub> represent a hydrogen atom, a methyl, or, considered jointly, they form a substituent =O, E represents -O- or -NG<sub>13</sub>-; A represents a C<sub>2</sub>-C<sub>6</sub> alkylene group or a -(CH<sub>2</sub>)<sub>3</sub>-O- group; x is 0 or 1; G<sub>13</sub> represents a hydrogen atom, a C<sub>1</sub>-C<sub>12</sub> alkyl group, a C<sub>2</sub>-C<sub>5</sub> hydroxyalkyl group, a C<sub>5</sub>-C<sub>7</sub> cycloalkyl group; G<sub>19</sub> has the same meanings as G<sub>18</sub> or it represents one of the following groups: -NG<sub>21</sub>G<sub>22</sub>, -OG<sub>23</sub>, -NHCH<sub>2</sub>OG<sub>23</sub>, or -N(CH<sub>2</sub>OG<sub>23</sub>)<sub>2</sub>; G<sub>20</sub>,

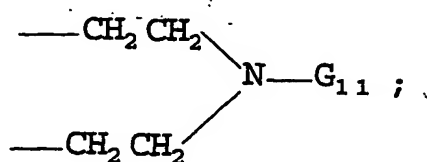
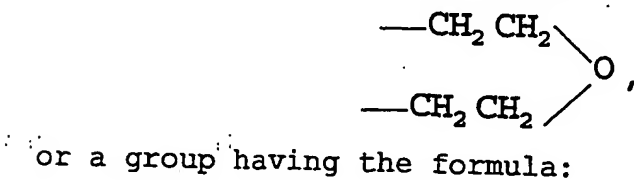
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when n is 1, has the same meanings as  $G_{18}$ , or  $G_{19}$ , if n is 2, it represents an -E-B-E- group wherein B represents a  $C_2-C_8$  alkylene group optionally interrupted by 1 or 2 -N( $G_{21}$ )- groups;  $G_{21}$  represents a  $C_1-C_{12}$  alkyl group, a cyclohexyl group, a benzyl, a  $C_1-C_4$  hydroxyalkyl group, or a group having the following general formula:



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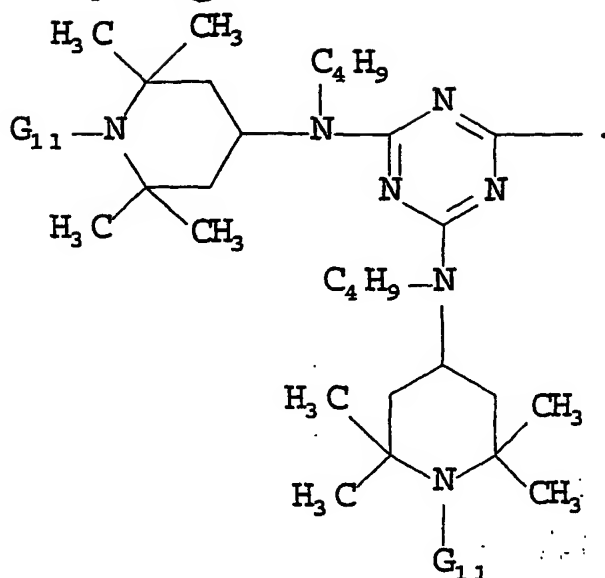
$G_{22}$  represents a  $C_1-C_{12}$  alkyl group, a cyclohexyl group, a benzyl, a  $C_1-C_4$  hydroxyalkyl group;  $G_{23}$  represents a hydrogen atom, a  $C_1-C_{12}$  alkyl group, a phenyl, or,  $G_{21}$  and  $G_{22}$ , considered jointly, represent a  $C_4-C_5$  alkylene or oxyalkylene group, for example:



20

$G_{21}$  is a group having the general formula:

25



5

Examples of  $C_1$ - $C_{12}$  alkyl groups are: methyl, ethyl, n-propyl, n-butyl, s-butyl, t-butyl, n-hexyl, n-octyl, 2-ethylhexyl, n-nonyl, n-decyl, n-undecyl, n-dodecyl, etc.

Examples of  $C_1$ - $C_4$  hydroxyalkyl groups are: 2-hydroxyethyl, 2-hydroxypropyl, 3-hydroxypropyl, 2-hydroxybutyl, 4-hydroxybutyl, etc.

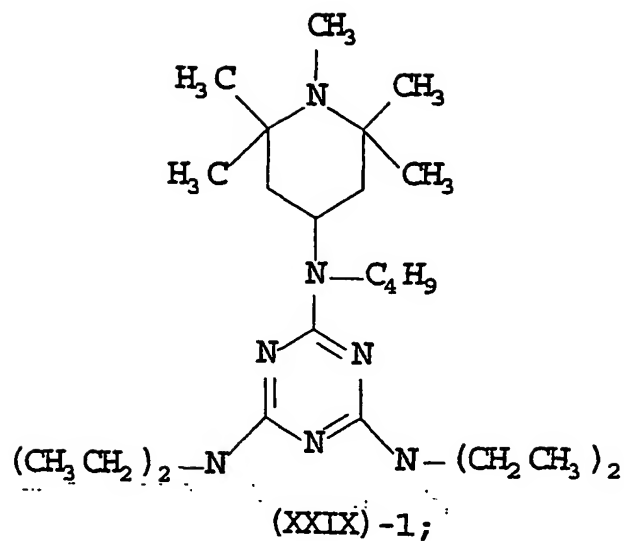
Examples of A, when it represents a  $C_2$ - $C_6$  alkylene group, are: ethylene, propylene, 2,2-dimethylpropylene, tetramethylene, hexamethylene, etc.

Examples of  $G_{21}$  and  $G_{22}$ , when, considered jointly, they represent a  $C_4$ - $C_5$  alkylene or oxyalkylene group, are: tetramethylene, pentamethylene, 3-oxapentamethylene, etc.

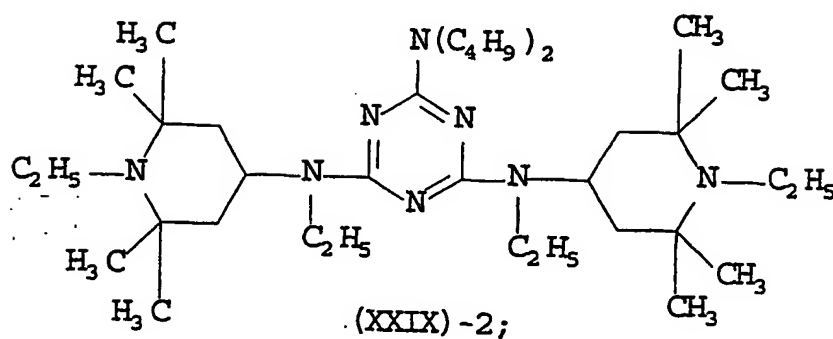
Specific examples of polyalkylpiperidines having general formula (XXIX) are:

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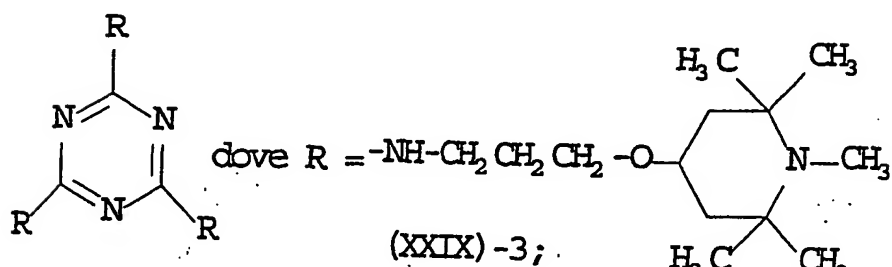
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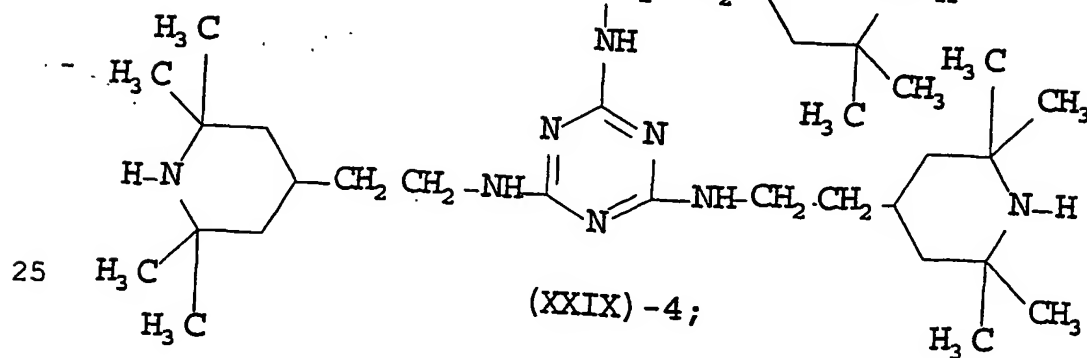
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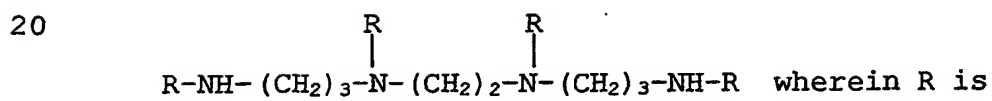
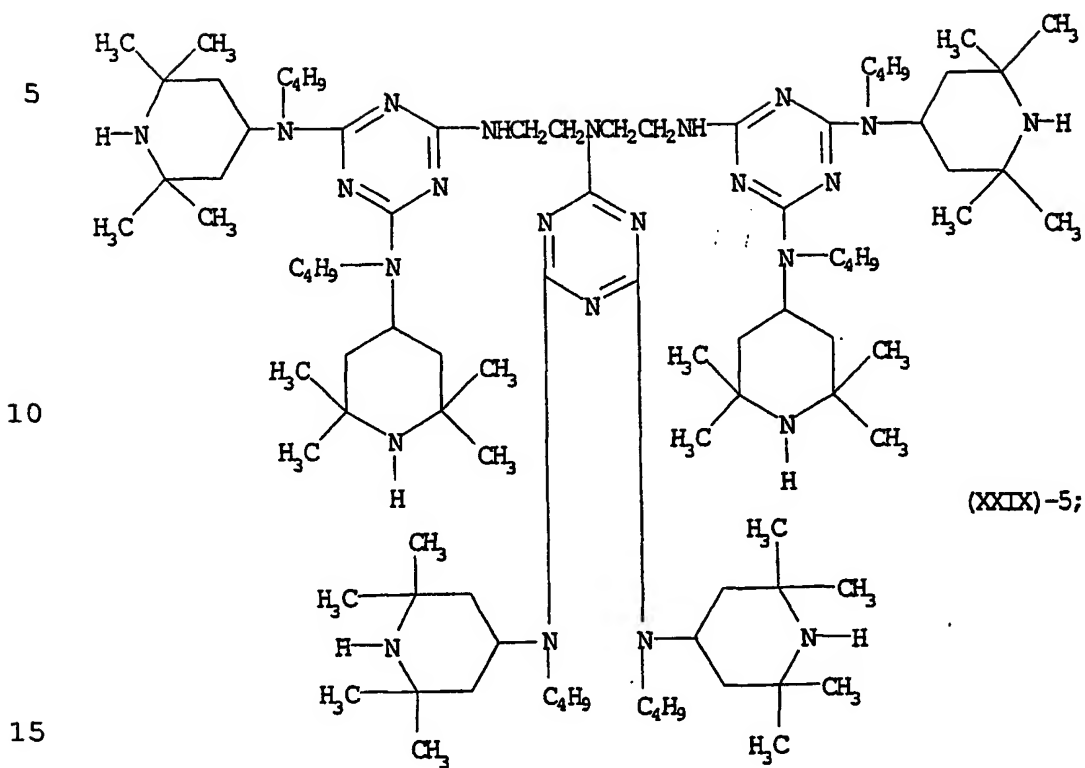
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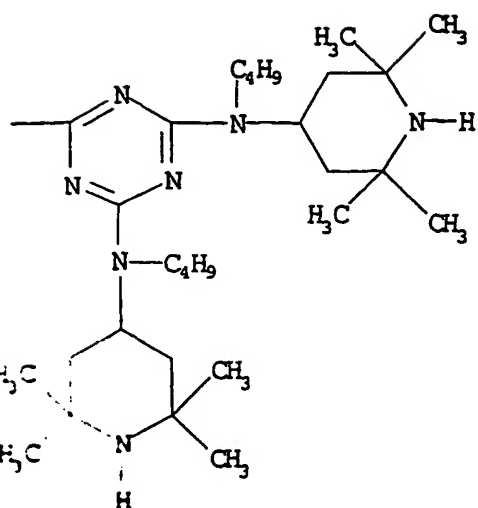
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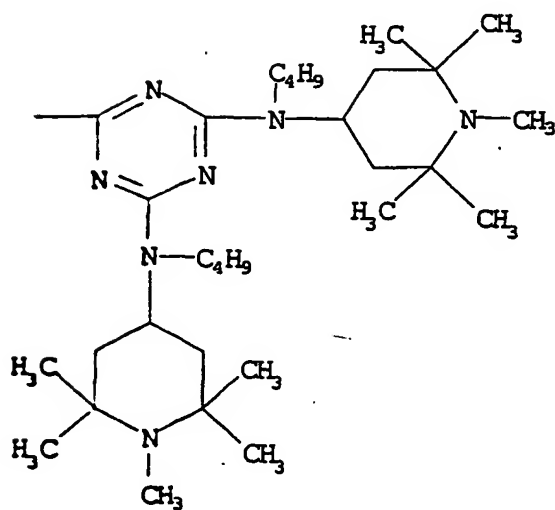
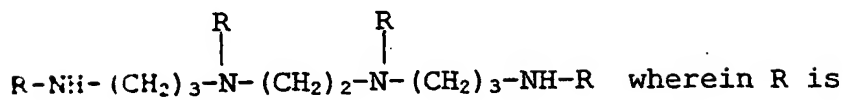
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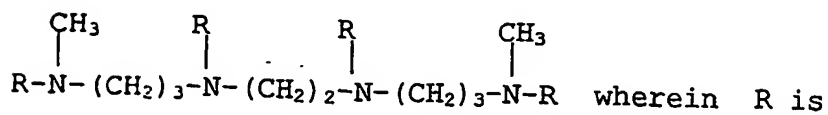




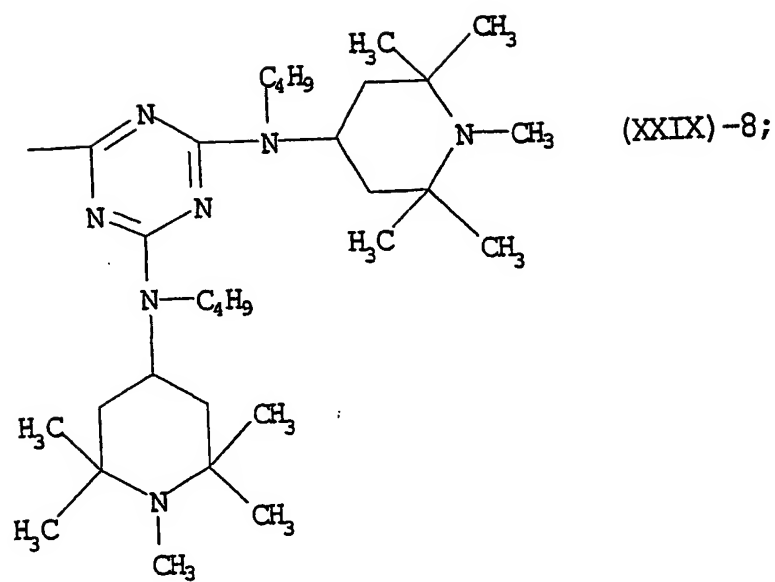
(XXIX)-6;



(XXIX)-7;



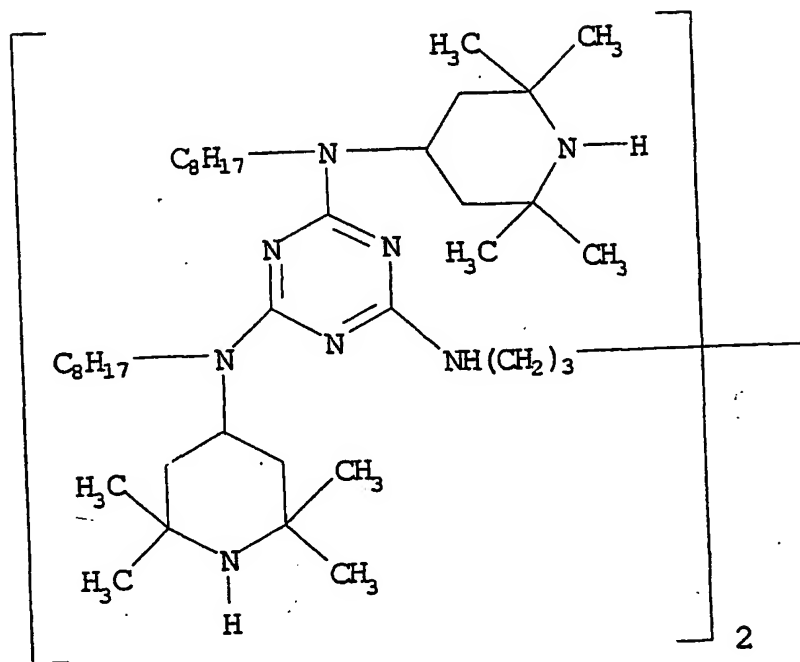
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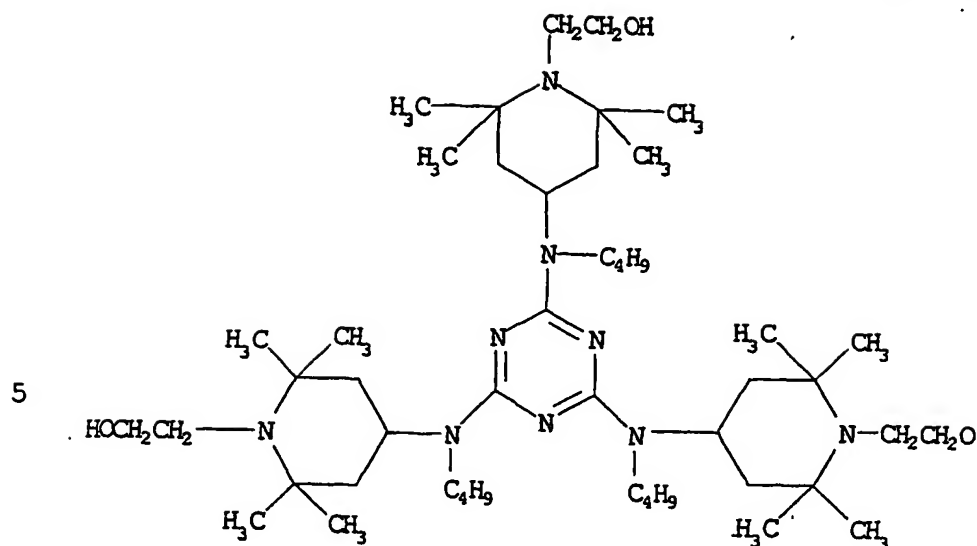
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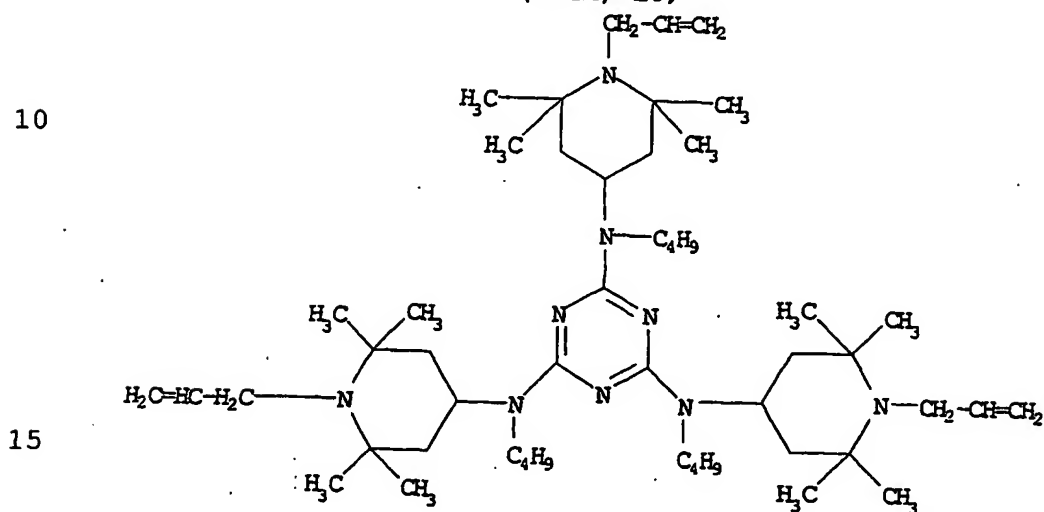


(XXIX)-9;

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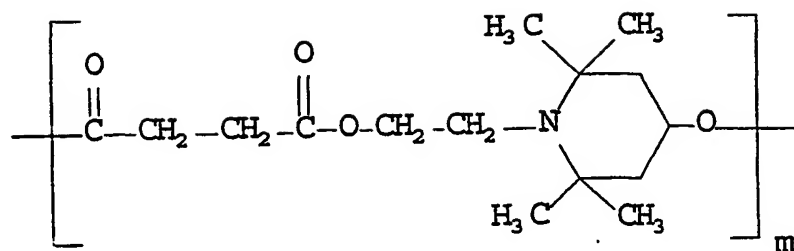
(XXIX)-10;



(XXIX)-11.

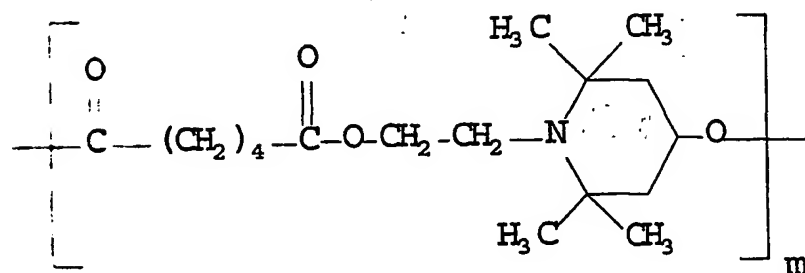
(f') Oligomeric or polymeric compounds whose recurrent structural unit contains a 2,2,6,6-tetramethylpiperidine radical, in particular polyesters, polyethers, polyamides, polyamines, polyurethanes, polyureas, polyaminotriazines, poly(meth)acrylates, poly(meth)acrylamides, and their copolymers containing said radical.

Specific examples of the above 2,2,6,6-polyalkylpiperidines are represented by the following formulae wherein m is a number ranging from 2 to 200:



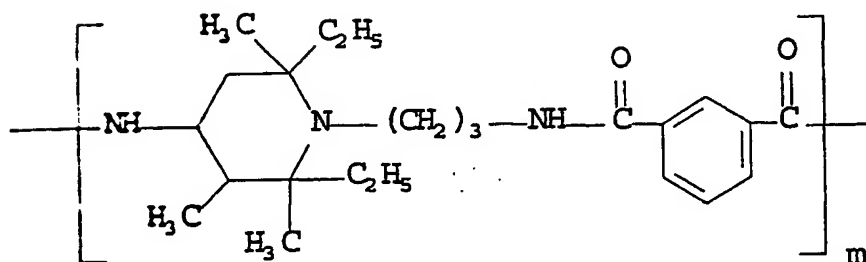
(XXX) -1;

5



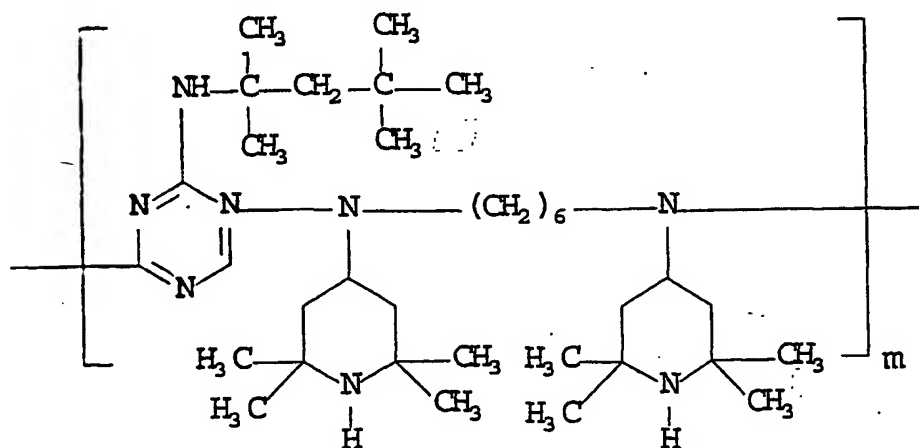
(XXX) -2;

10



(XXX) -3;

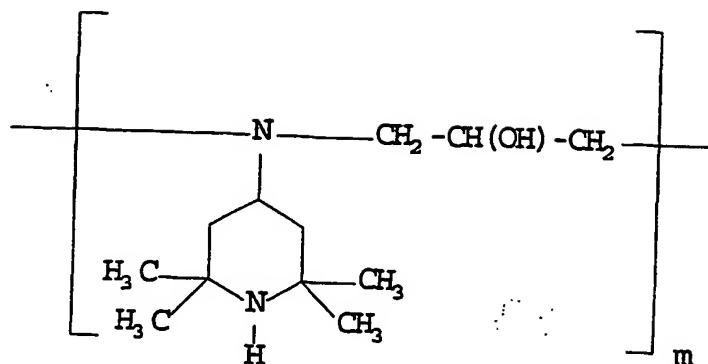
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(XXX) -4;

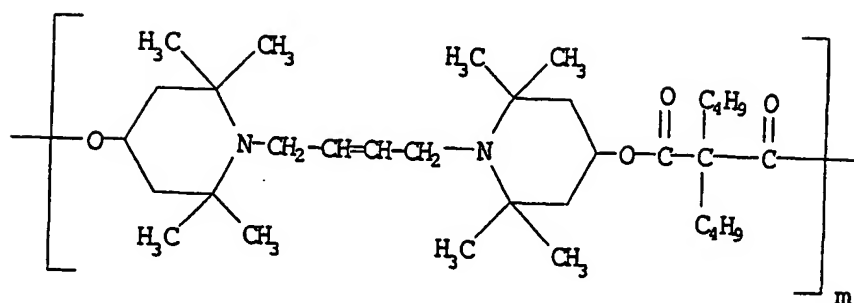
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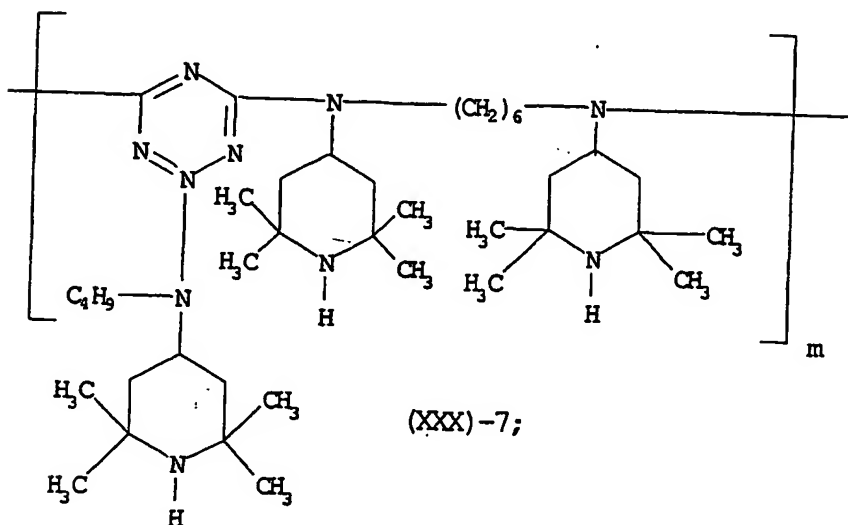
(XXX)-5;

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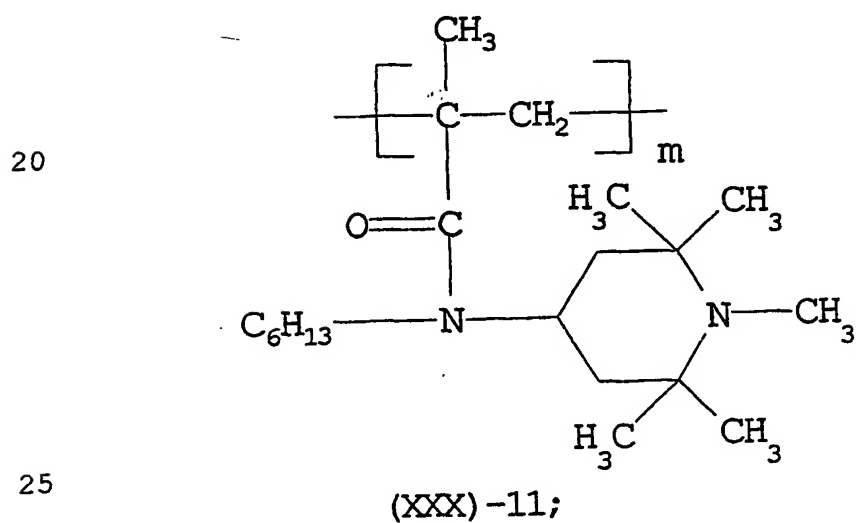
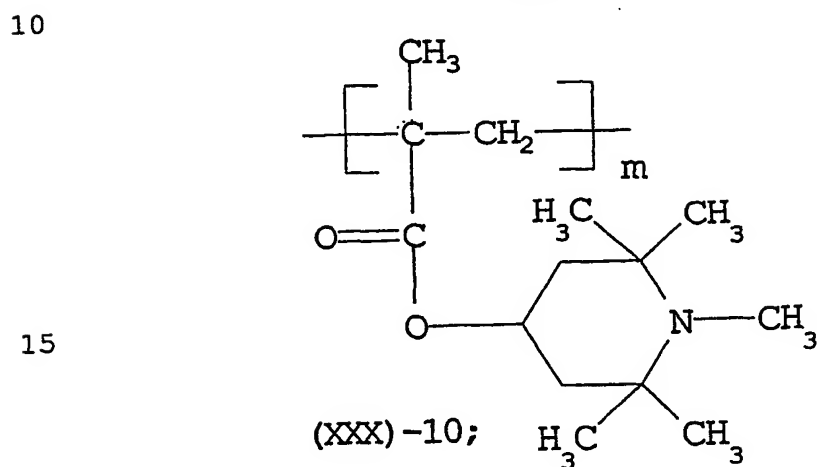
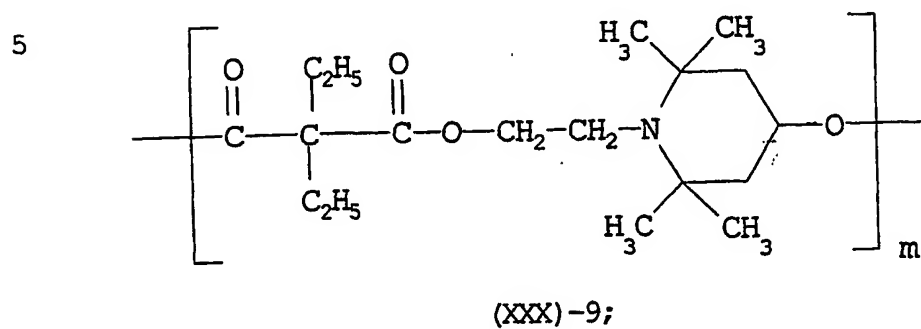
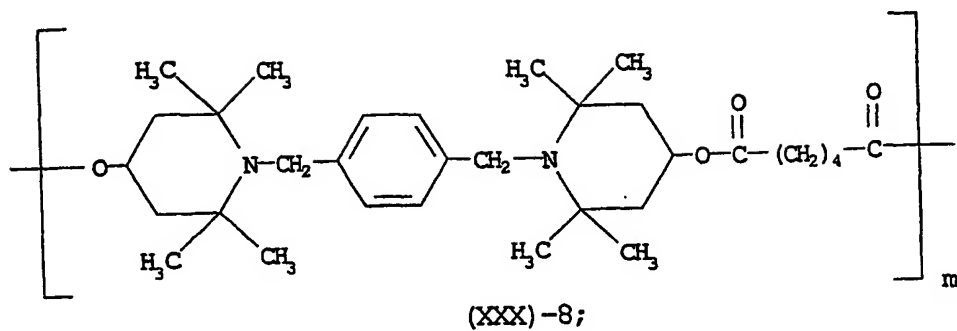
(XXX)-6;

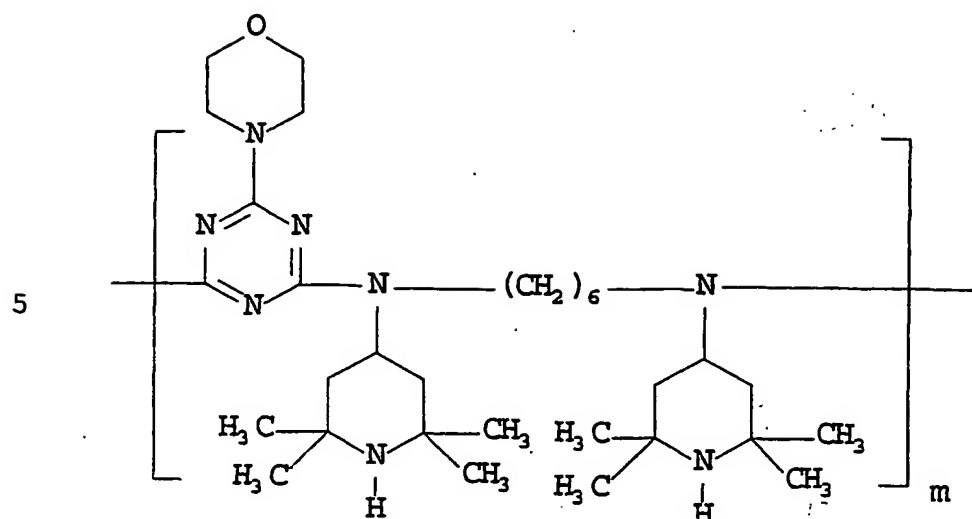
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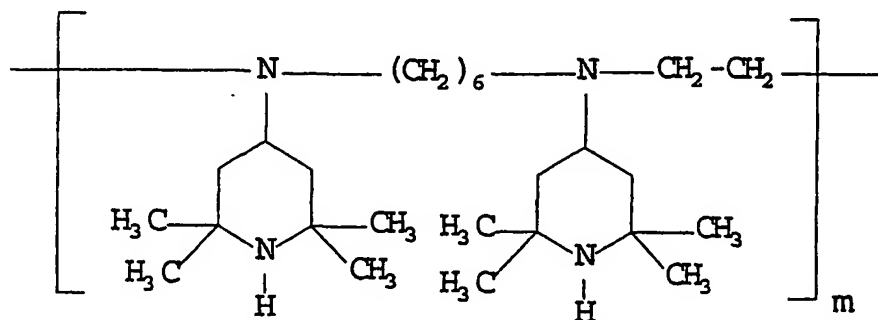
(XXX)-7;

25

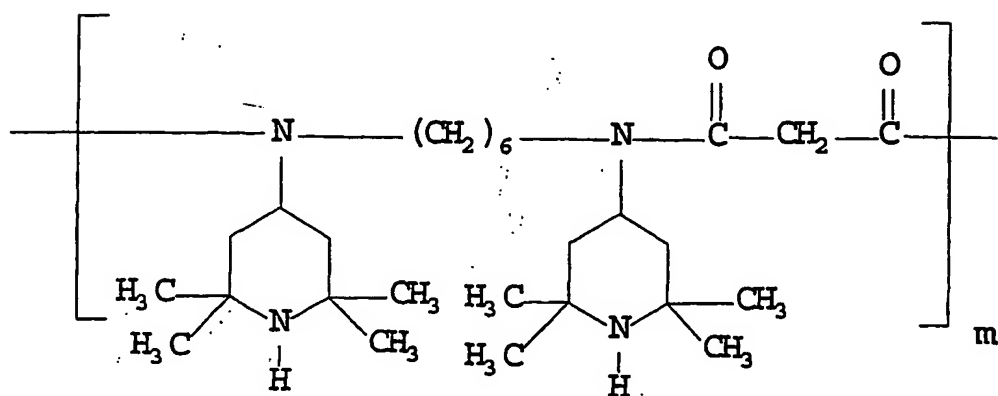




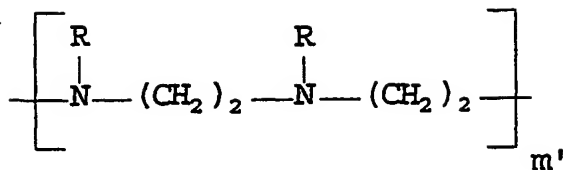
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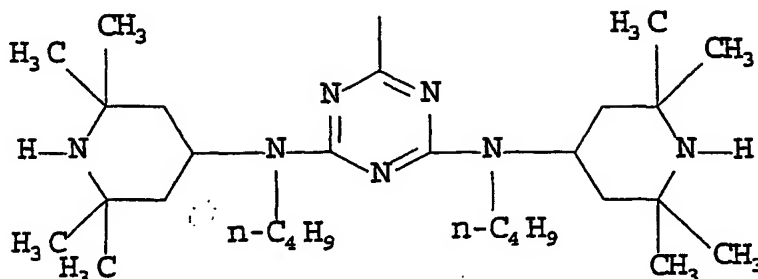


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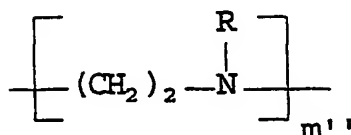


wherein R =

5



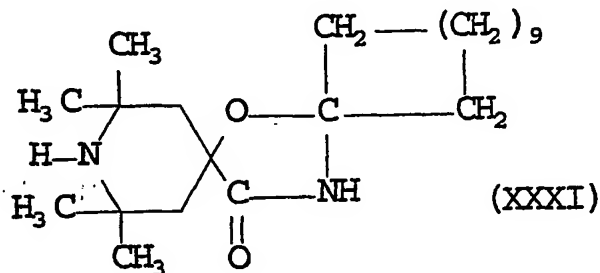
or R =



10 wherein  $m'$  and  $m''$  are an integer ranging from 0 to 200, extremes included, on the condition that  $m' + m''$  is  $m$ .

Further examples of polymeric compounds ( $f'$ ) useful for the purposes of the present invention are:

15 - the reaction products between compounds having formula (XXXI):



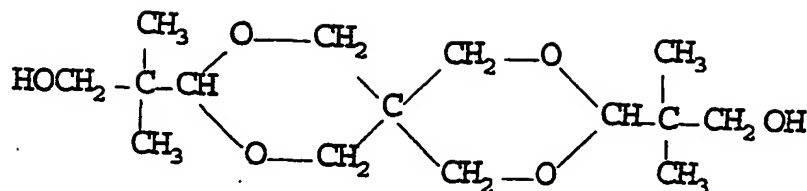
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and epichlorohydrin;

- polyesters obtained from the reaction of butane-1,2,3,4-tetracarboxylic acid with a bifunctional alcohol having formula (XXXII):

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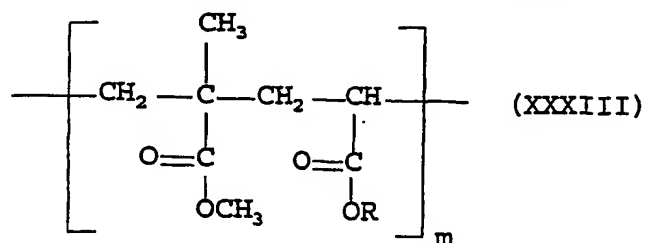




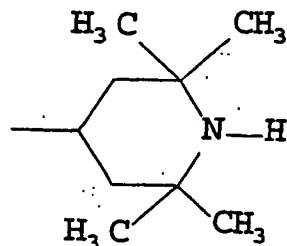
(XXXII)

whose carboxylic termination deriving from tetracar-  
boxylic acid has been esterified with a 2,2,6,6-  
tetramethyl-4-hydroxypiperidine group;

compounds having general formula (XXXIII):

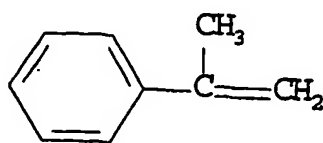


wherein about a third of the R radicals represent a  
-C<sub>2</sub>H<sub>5</sub> group and the remainder a group having the  
formula:

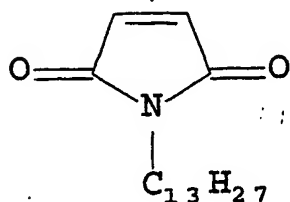


and *m* is a number ranging from 2 to 200, extremes  
included;

copolymers whose recurrent unit consists of two  
units having the formula:

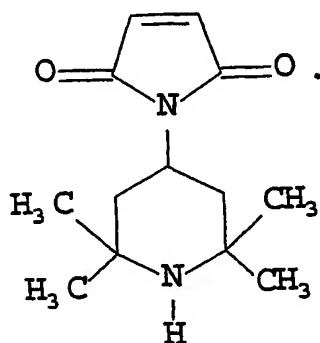


and, in each case, of a unit having the formula:



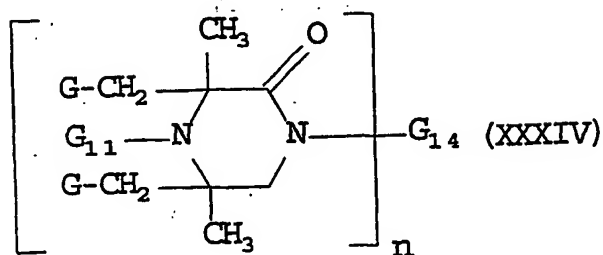
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and a unit having the formula:



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(g') Compounds having general formula (XXXIV):



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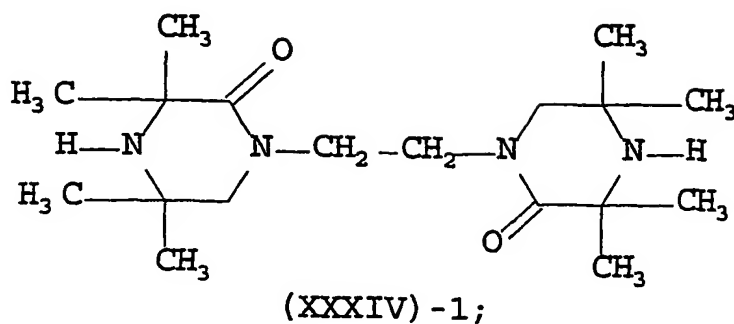
wherein  $n$  is 1 or 2,  $G$  and  $G_{11}$  have the same meanings defined above under point (a') and  $G_{14}$  has the same meanings described above under point (b'), on the condition that  $G_{14}$  can never represent the group  $-\text{CONH}-Z$  or the group  $-\text{CH}_2-\text{CH}(\text{OH})-\text{CH}_2-\text{O}-D-\text{O}$ .

Examples of compounds having general formula (XXXIV)

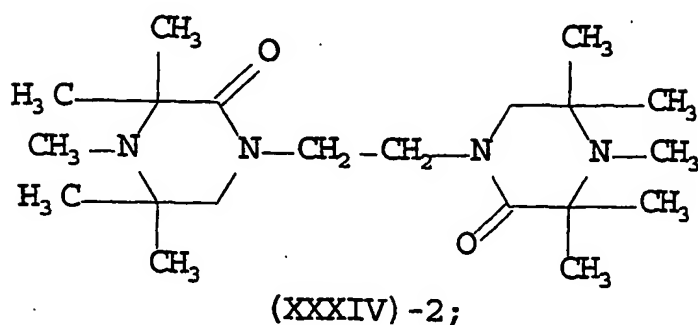
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are the following:

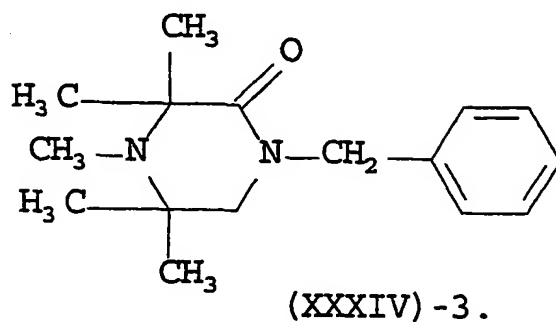
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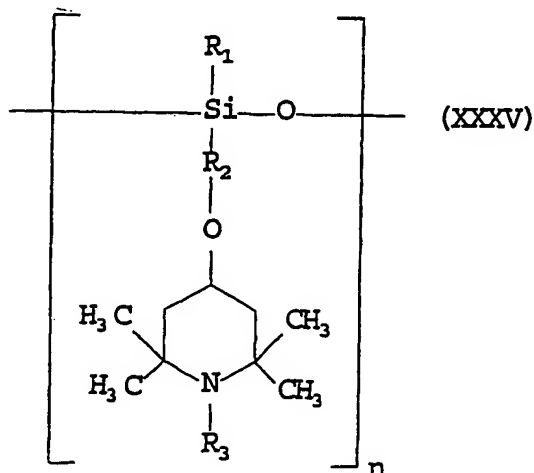


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(h') Compounds having general formula (XXXV):

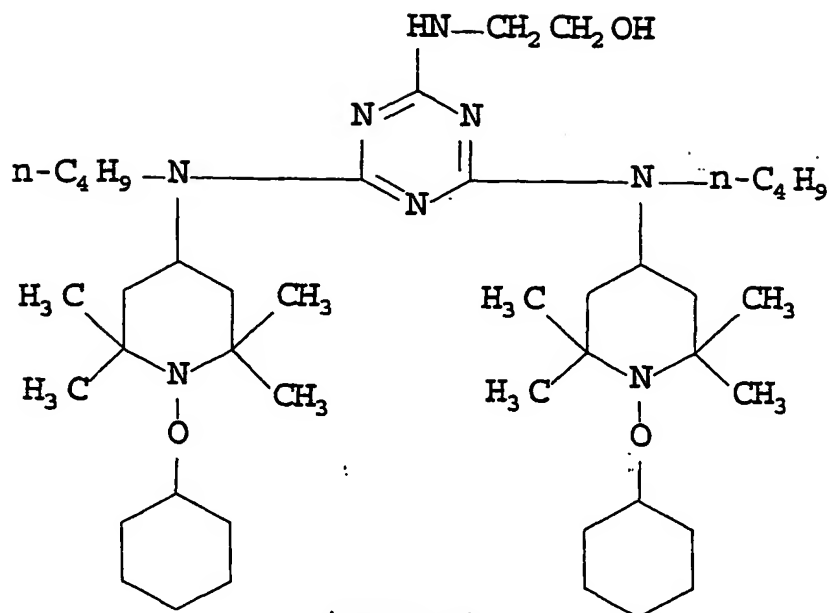
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25

wherein  $R_1$  represents a  $C_1$ - $C_{10}$  alkyl group, a  $C_5$ - $C_{12}$  cyclo-  
alkyl group optionally substituted with a  $C_1$ - $C_4$  alkyl  
group, a phenyl optionally substituted with a  $C_1$ - $C_{10}$  alkyl  
5 group;  $R_2$  represents a  $C_3$ - $C_{10}$  alkylene group;  $R_3$  repre-  
sents a hydrogen atom, a  $C_1$ - $C_8$  alkyl group, O, a -  
CH<sub>2</sub>CN group, a  $C_3$ - $C_6$  alkenyl group, a  $C_7$ - $C_9$  phenylalkyl  
group optionally substituted in the phenyl radical with a  
 $C_1$ - $C_4$  alkyl group, a  $C_1$ - $C_8$  acyl group, an -OR'<sub>3</sub> group  
10 wherein R'<sub>3</sub> represents a  $C_1$ - $C_{10}$  alkyl group; and n is a  
number ranging from 1 to 50, extremes included.

Of particular interest for the purposes of the pres-  
ent invention are compounds belonging to the group of  
sterically hindered amines (d), selected from: Tinuvin®  
15 123 of Ciba Specialty Chemicals; Tinuvin® 144 of Ciba  
Specialty Chemicals; Lowilite® 76 of Great Lakes Chemical  
Corporation; Lowilite® 62 of Great Lakes Chemical Corpo-  
ration; Lowilite® 94 of Great Lakes Chemical Corporation;  
Chimassorb® 119 of Ciba Specialty Chemicals; the compound  
20 having formula (XXXV)-1:



10 poly-methylpropyl-3-oxy-[4-(2,2,6,6-tetramethyl)piperid-  
inyl]siloxane, known under the trade-name of UVASIL<sup>®</sup> 299  
of Great Lakes Chemical Corporation; polymethylpropyl-3-  
oxy-[4-(1,2,2,6,6-pentamethyl)piperidiny]siloxane.

15 Compounds belonging to the group of sterically hin-  
dered amines (d) useful for the purposes of the present  
invention are those having an average molecular weight  $M_n$   
ranging from 500 to 10,000, in particular from 1,000 to  
10,000.

20 The stabilizing mixtures, object of the present in-  
vention, are capable of stabilizing organic polymers  
against degradation caused by oxygen, heat and/or light.  
Examples of organic polymers to which they can be added  
are:

25 1. Polymers of mono-olefins and di-olefins such as, for

example, polypropylene, polyisobutylene, polybut-1-ene, poly-4-methylpent-1-ene, polyisoprene or polybutadiene; as well as polymers of cyclo-olefins such as, for example, cyclopentene or norbornene; polyethylene (which can be optionally cross-linked) such as, for example, high density polyethylene (HDPE), high density and high molecular weight polyethylene (HDPE-HMW), high density and ultrahigh molecular weight polyethylene (HDPE-UHMW), medium density polyethylene (MDPE), low density polyethylene (LDPE), linear low density polyethylene (LLDPE), branched low density polyethylene (BLDPE), (VLDPE), (ULDPE).

Polyolefins such as, for example the mono-olefins mentioned in the above paragraph, preferably polyethylene and polypropylene, can be prepared with many methods known in literature, preferably using the following methods:

- (a) radicalic polymerization (generally carried out at a high pressure and high temperature);
- (b) catalytic polymerization using a catalyst which normally contains one or more metals of groups IVb, Vb, VIb or VIII of the Periodic Table. These metals generally have one or more ligands such as, for example, oxides, halides, alcoholates, esters, ethers, amines, alkyls, alkenyls and/or aryls which can be  $\pi$ - or  $\sigma$ -coordinated. These metal complexes can be in free form or supported on substrates such as, for example,

activated magnesium chloride, titanium(III) chloride, alumina or silicon oxide. Said catalysts can be soluble or insoluble in the polymerization medium. The catalysts can be used alone or in the presence of other activators such as, for example, metal alkyls, metal hydrides, halides of metal alkyls, oxides of metal alkyls or metal alkyloxanes, these metals being elements belonging to groups Ia, IIa and/or IIIa of the Periodic Table. The activators can be conveniently modified with other ester, ether, amine or silyl-ether groups. These catalytic systems are usually called Phillips, Standard Oil Indiana, Ziegler(-Natta), TNZ (Du-Pont), metallocene or "single site catalyst" (SSC).

2. Mixtures of the polymers described under point (1) such as, for example, mixtures of polypropylene with polyisobutylene; mixtures of polypropylene with polyethylene (for example, PP/HDPE, PP/LDPE); mixtures of different types of polyethylene (for example, LDPE/HDPE).
3. Copolymers of mono-olefins and di-olefins with each other or with other vinyl monomers such as, for example, ethylene/propylene copolymers, linear low density polyethylene (LLDPE) and its mixtures with low density polyethylene (LDPE), propylene/but-1-ene copolymers, propylene/isobutylene copolymers, ethylene/but-1-ene copolymers,

ethylene/hexene copolymers, ethylene/methylpentene copolymers, ethylene/heptene copolymers, ethylene/octene copolymers, propylene/butadiene copolymers, isobutylene/isoprene copolymers, ethylene/alkyl acrylate copolymers, ethylene/alkyl methacrylate copolymers, ethylene/vinyl acetate copolymers and their copolymers with carbon monoxide or ethylene/acrylic acid copolymers and their salts (ionomers) as well as terpolymers of ethylene with propylene and a diene such as, for example, hexadiene, dicyclopentadiene or ethylidene-norbornene; and also mixtures of said copolymers with each other or with the polymers cited in under point (1) such as, for example, polypropylene/ethylene/propylene copolymers, LDPE/ethylene/vinyl acetate (EVA) copolymers, LDPE/ethylene/acrylic acid (EAA) copolymers, LLDPE/EVA, LLDPE/EAA, and alternating or random polyalkylene/carbon monoxide copolymers and their mixtures with other polymers such as, for example, polyamides.

4. Hydrocarbon resins (for example, C<sub>5</sub>-C<sub>9</sub>) comprising their hydrogenated modifications (for example, adhesive resins) and mixtures with polyalkylene and starch.

5. Polystyrene, poly(p-methylstyrene), poly( $\alpha$ -methylstyrene).

6. Copolymers of styrene or  $\alpha$ -methylstyrene with dienes or acrylic derivatives such as, for example, styrene-



ne/butadiene, styrene/acrylonitrile, styrene/alkyl methacrylate, styrene/butadiene/alkyl acrylate, styrene/butadiene/alkyl methacrylate, styrene/maleic anhydride, styrene/acrylonitrile/methyl acrylate; mixtures, having a high impact strength, between copolymers of styrene and another polymer such as, for example, a polyacrylate, a polymer of a diene or an ethylene/propylene/diene terpolymer, block copolymers of styrene such as, for example, styrene/butadiene/styrene, styrene/isoprene/styrene, styrene/ethylene/butylene/styrene or styrene/ethylene/propylene/styrene.

7. Grafted copolymers of styrene or of  $\alpha$ -methylstyrene such as, for example, styrene in polybutadiene, styrene in polybutadiene/styrene or polybutadiene/acrylonitrile copolymers; styrene and acrylonitrile (or methacrylonitrile) in polybutadiene; styrene, acrylonitrile and methylmethacrylate in polybutadiene; styrene and maleic anhydride in polybutadiene; styrene, acrylonitrile and maleic anhydride or maleimide in polybutadiene; styrene and maleimide in polybutadiene; styrene and alkylacrylates or alkylmethacrylates in polybutadiene; styrene and acrylonitrile in ethylene/propylene/diene terpolymers, styrene and acrylonitrile in polyalkyl acrylates or polyalkyl methacrylates, styrene and acrylonitrile in acrylate/butadiene copolymers, as well as mixtures of the

copolymers listed above with the copolymers cited under point (6) such as, for example, mixtures of known copolymers such as ABS, MBS, ASA or AES.

8. Polymers containing halogens such as, for example,  
5 polychloroprene, chlorinated rubbers, chlorinated or brominated isobutylene-isoprene copolymers ("halobutyl rubber"), chlorinated or chlorosulfonated polyethylene, ethylene and chlorinated ethylene copolymers, homopolymers and copolymers of epichlorohydrin, in particular polymers  
10 of vinyl compounds containing halogens such as, for example, polyvinyl chloride, polyvinylidene chloride, polyvinyl fluoride or polyvinylidene fluoride; and also their copolymers such as, for example, vinyl chloride/vinylidene chloride, vinyl chloride/vinyl acetate or vinylidene chloride/vinyl acetate.  
15

9. Polymers deriving from  $\alpha,\beta$ -unsaturated acids and their derivatives such as, for example, polyacrylates and polymethacrylates, polymethyl methacrylates, polyacrylamides and polyacrylonitriles, impact modified with butyl  
20 acrylate.

10. Copolymers of monomers according to point (9) with each other or with other unsaturated monomers such as, for example, acrylonitrile/butadiene copolymers, acrylonitrile/alkyl acrylate copolymers, acrylonitrile/alkoxyalkyl  
25 acrylate copolymers or acrylonitrile/vinyl halide copoly-

mers or acrylonitrile/alkyl methacrylate/butadiene terpolymers.

11. Polymers deriving from unsaturated alcohols and amines, or their acyl or acetal derivatives such as, for example, polyvinyl alcohol, polyvinyl acetate, polyvinyl stearate, polyvinyl benzoate, polyvinyl maleate, polyvinyl butyral, polyallyl phthalate or polyallyl melamine; and also their copolymers with the olefins listed under point (1).
12. Homopolymers and copolymers of open-chain ethers or cyclic ethers such as, for example, polyalkylene glycols, polyethylene oxide, polypropylene oxide, or copolymers of the compounds described above with bis-glycidyl ethers.
13. Polyacetals such as, for example, polyoxymethylene and those polyoxymethylenes containing comonomers, for example, ethylene oxide; polyacetals modified with thermoplastic polyurethanes, acrylates or MBS.
14. Polyphenylene oxides and sulfides and their mixtures with styrene polymers or polyamides.
15. Polyurethanes deriving from hydroxyl-terminated polyethers, polyesters or polybutadienes on the one hand and aliphatic or aromatic polyisocyanates on the other, as well as their precursors.
16. Polyamides and copolyamides deriving from diamines and dicarboxylic acids and/or aminocarboxylic acids or

from the corresponding lactams such as, for example, polyamide 4, polyamide 6, polyamide 6/6, 6/10, 6/9, 6/12, 4/6, 12/12, polyamide 11, polyamide 12, aromatic polyamides obtained starting from m-xylene diamine and adipic acid; 5 polyamides prepared from hexamethylenediamine and isophthalic and/or terephthalic acid and with or without an elastomer as modifier, for example, poly-2,4,4-trimethylhexamethylene terephthalamide or poly-m-phenylene isophthalamide; and also block copolymers of the above 10 polyamides with polyolefins, olefinic copolymers, ionomers or elastomers chemically bound or grafted; or with polyethers such as, for example, polyethylene glycol, polypropylene glycol or polytetramethylene glycol; as well as polyamides or copolyamides modified with EPDM or ABS; and 15 polyamides condensed during processing ("RIM polyamide system").

17. Polyureas, polyimides, polyamide-imides, polyetherimides, polyesterimides, polyhydantoins, and polybenzimidazoles.

20 18. Polyesters deriving from dicarboxylic acids and diols and/or from hydroxycarboxylic acids or from the corresponding lactones such as, for example, polyethylene terephthalate, polybutylene terephthalate, poly-1,4-dimethylolcyclohexane terephthalate and polyhydroxybenzoates, 25 as well as block copolyether esters deriving from polyeth-

ers with hydroxyl-terminated groups; and also polyesters modified with polycarbonates or MBS.

19. Polycarbonates and polyester carbonates.
20. Polysulfones, polyethersulfones and polyetherketones.
- 5 21. Cross-linked polymers deriving from aldehydes on the one hand and from phenols, urea and melamines on the other, such as, for example, phenol/formaldehyde resins, urea/formaldehyde resins and melamine/formaldehyde resins.
- 10 22. Dried or non-dried alkyd resins.
23. Resins based on unsaturated polyesters deriving from copolyesters of dicarboxylic acids saturated and unsaturated with polyhydric alcohols and vinyl compounds as cross-linking agents, and also the above resins containing
- 15 halogens and having a good flame-resistance.
24. Cross-linkable acrylic resins deriving from substituted acrylates such as, for example, epoxy acrylates, urethane acrylates or polyester acrylates.
25. Alkyd resins, resins based on polyesters or acrylated
- 20 resins cross-linked with melamine resins, resins based on urea, resins based on isocyanates, resins based on isocyanurates, resins based on polyisocyanates or epoxy resins.
26. Cross-linked epoxy resins deriving from aliphatic,
- 25 cycloaliphatic, heterocyclic or aromatic glycidyl com-

pounds such as, for example, products of diglycidyl ethers of bisphenol A and bisphenol F, which are cross-linked with the usual cross-linking agents such as, for example, anhydrides or amines, in the presence of or without accelerating agents.

27. Natural polymers such as, for example, cellulose, natural rubber, gelatin; and their derivatives chemically modified to give homologous polymers such as, for example, cellulose acetates, propionates and butyrates, or cellulose ethers such as methyl-cellulose; as well as hydrocarbon resins ("rosins") and their derivatives.

28. Mixtures of the above polymers ("polyblends") such as, for example, PP/EPDM, polyamides/EPDM or ABS, PVC/EVA, PVC/ABS, PVC/MBS, PC/ABS, PBTP/ABS, PC/ASA, PC/PBT, PVC/CPE, PVC/acrylates, POM/thermoplastic PUR, PC/thermoplastic PUR, POM/acrylates, POM/MBS, PPO/HIPS, PPO/PA 6.6 and copolymers, PA/HDPE, PA/PP, PA/PPO, PBT/PC/ABS, PBT/PET/PC.

29. Natural or synthetic organic materials which are pure monomeric compounds or mixtures of said compounds, such as, for example, mineral oils, animal or vegetable oils, fats or waxes, oils, fats or waxes based on synthetic esters (for example, phthalates, adipates, phosphates, trimellitates), as well as mixtures of synthetic esters with mineral oils in any weight ratio, in particu-

lar those used in spinning compositions, as well as aqueous emulsions of said organic materials.

30. Aqueous emulsions of natural or synthetic rubbers such as, for example, natural latex or latexes based on  
5 carboxylated styrene-butadiene copolymers.

The organic polymers which can be stabilized with the mixtures, object of the present invention, are, preferably, natural, semi-synthetic or synthetic polymers selected from those described above. More preferably, the  
10 mixtures, object of the present invention, are useful in the stabilization of thermoplastic polymers, especially polyolefins, in particular polyethylene and polypropylene or their copolymers with mono- and di-olefins.

A further object of the present invention therefore  
15 relates to polymeric compositions containing an organic polymer and an effective quantity of one of the stabilizing mixtures, object of the present invention. Yet another object of the present invention relates to the end-products obtained from the processing of the above poly-  
20 meric compositions.

The stabilizing mixtures, object of the present invention, are particularly useful against degradation caused by oxygen and heat and are consequently exceptionally useful as process stabilizers.

25 Compounds (a), (b) and (c) and, optionally, (d) of

the above stabilizing mixtures can be added to the organic polymers to be stabilized either individually or mixed with each other.

Compound (a) is added to the organic polymers to be stabilized in a quantity ranging from 0.001% to 2.5% with respect to the weight of the organic polymer to be stabilized, preferably from 0.005% to 2%, for example, from 0.01% to 1%.

Compounds (b), (c) and, optionally, (d), are added to the organic polymers to be stabilized in a quantity ranging from 0.01% to 10% with respect to the weight of the organic polymer to be stabilized, for example from 0.01% to 5%, preferably from 0.025% to 3%, even more preferably from 0.025% to 1%.

The stabilizing mixtures object of the present invention can optionally contain other stabilizers (co-stabilizers).

Stabilizers for organic polymers useful for the purpose are selected from the following groups:

1. Antioxidants

1.1 Alkylated monophenols such as, for example: 2,6-di-t-butyl-4-methylphenol; 2-t-butyl-4,6-dimethylphenol; 2,6-di-t-butyl-4-ethylphenol; 2,6-di-t-butyl-4-n-butylphenol; 2,6-di-t-butyl-4-isobutylphenol; 2,6-dicyclopentyl-4-methylphenol; 2-( $\alpha$ -methylcyclohexyl)-4,6-dime-



thylphenol; 2,6-dioctadecyl-4-methylphenol; 2,4,6-tri-cyclohexylphenol; 2,6-di-t-butyl-4-methoxymethylphenol; nonylphenols with a linear or branched alkyl chain such as, for example, 2,6-dinonyl-4-methylphenol; 2,4-dimethyl-6-(1'-methylundec-1'-yl)phenol; 2,4-dimethyl-6-(1'-methylheptadec-1'-yl)phenol; 2,4-dimethyl-6-(1'-methyltridec-1'-yl)phenol; and their mixtures.

1.2 Alkylthiomethylphenols such as, for example: 2,4-dioctylthiomethyl-6-t-butylphenol; 2,4-dioctylthiomethyl-6-methylphenol; 2,4-dioctylthiomethyl-6-ethylphenol; 2,6-didodecylthiomethyl-4-nonylphenol.

1.3 Hydroquinones and alkylated hydroquinones such as, for example: 2,6-di-t-butyl-4-methoxyphenol; 2,5-di-t-butylhydroquinone; 2,5-di-t-amylhydroquinone; 2,6-diphenyl-4-octadecyloxyphenol; 2,6-di-t-butylhydroquinone; 2,5-di-t-butyl-4-hydroxyanisol; 3,5-di-t-butyl-4-hydroxyanisol; 3,5-di-t-butyl-4-hydroxyphenyl stearate; bis(3,5-di-t-butyl-4-hydroxyphenyl)adipate.

1.4 Tocopherols such as, for example:  $\alpha$ -tocopherol,  $\beta$ -tocopherol,  $\gamma$ -tocopherol,  $\delta$ -tocopherol and their mixtures (Vitamin E).

1.5 Hydroxylated thiodiphenyl ethers such as, for example 2,2'-thiobis-(6-t-butyl-4-methylphenol); 2,2'-thiobis-(4-octylphenol); 4,4'-thiobis-(6-t-butyl-3-methylphenol); 4,4'-thiobis-(6-t-butyl-2-methylphenol); 4,4'-thiobis-

(3,6- di-s-amylphenol); 4,4'-bis-(2,6-dimethyl-4-hydroxyphenyl)disulfide.

1.6 Alkylidene-bisphenols such as, for example: 2,2'-methylenebis-(6-t-butyl-4-methylphenol); 2,2'-methylenebis-(6-t-butyl-4-ethylphenol); 2,2'-methylenebis[4-methyl-6-( $\alpha$ -methylcyclohexyl)phenol]; 2,2'-methylenebis(4-methyl-6-cyclohexylphenol); 2,2'-methylenebis(6-nonyl-4-methylphenol); 2,2'-methylenebis(4,6-di-t-butylphenol); 2,2'-ethylidenebis(4,6-di-t-butylphenol); 2,2'-ethylidenebis(6-t-butyl-4-isobutylphenol); 2,2'-methylenebis[6-( $\alpha$ -methylbenzyl)-4-nonylphenol]; 2,2'-methylenebis[6-( $\alpha,\alpha$ -dimethylbenzyl)-4-nonylphenol]; 4,4'-methylenebis(2,6-di-t-butylphenol); 4,4'-methylenebis(6-t-butyl-2-methylphenol); 1,1-bis(5-t-butyl-4-hydroxy-2-methylphenyl)butane; 2,6-bis(3-t-butyl-5-methyl-2-hydroxybenzyl)-4-methylphenol; 1,1,3-tris-(5-t-butyl-4-hydroxy-2-methylphenyl)butane; 1,1-bis(5-t-butyl-4-hydroxy-2-methylphenyl)-3-n-dodecylmercaptobutane; ethyleneglycol bis[3,3-bis-(3'-t-butyl-4'-hydroxyphenyl)butyrate]; bis-(3-t-butyl-4-hydroxy-5-methylphenyl)dicyclopentadiene; bis[2-(3'-t-butyl-2'-hydroxy-5'-methylbenzyl)-6-t-butyl-4-methylphenyl]terephthalate; 1,1-bis(3,5-dimethyl-2-hydroxyphenyl)butane; 2,2-bis(3,5-di-t-butyl-4-hydroxyphenyl)propane; 2,2-bis(5-t-butyl-4-hydroxy-2-methylphenyl)-4-n-dodecylmercaptobutane; 1,1,5,5-tetra(5-t-butyl-4-hydroxy-

2-methylphenyl)pentane.

1.7 Benzyl compounds containing O, N or S such as, for example: 3,5,3',5'-tetra-*t*-butyl-4,4'-dihydroxydibenzyl-ether; octadecyl-4-hydroxy-3,5-dimethylbenzylmercapto-  
5 acetate; tridecyl-4-hydroxy-3,5-di-*t*-butyl-benzylmercaptoacetate; tris(3,5-di-*t*-butyl-4-hydroxybenzyl)amine; bis(4-*t*-butyl-3-hydroxy-2,6-dimethylbenzyl)dithioterephthalate; bis(3,5-di-*t*-butyl-4-hydroxybenzyl)sulfide; iso-octyl-3,5-di-*t*-butyl-4-hydroxybenzylmercaptoacetate.

10 1.8 Hydroxybenzylated malonates such as, for example: dioctadecyl-2,2-bis(3,5-di-*t*-butyl-2-hydroxybenzyl)malonate; dioctadecyl-2-(3-*t*-butyl-4-hydroxy-5-methylbenzyl)-malonate; didodecylmercaptoethyl-2,2-bis(3,5-di-*t*-butyl-4-hydroxybenzyl)malonate; bis[4-(1,1,3,3-tetramethyl-  
15 butyl)phenyl]-2,2-bis(3,5-di-*t*-butyl-4-hydroxybenzyl)-malonate.

1.9 Aromatic hydroxybenzyl compounds such as, for example: 1,3,5-tris(3,5-di-*t*-butyl-4-hydroxybenzyl)-2,4,6-trimethylbenzene; 1,4-bis(3,5-di-*t*-butylhydroxybenzyl)-  
20 2,3,5,6-tetramethylbenzene; 2,4,6-tris(3,5-di-*t*-butyl-4-hydroxybenzyl)phenol.

1.10 Triazine compounds such as, for example: 2,4-bis(octylmercapto)-6-(3,5-di-*t*-butyl-4-hydroxyaniline)-1,3,5-triazine; 2-octylmercapto-4,6-bis(3,5-di-*t*-butyl-4-  
25 hydroxyaniline)-1,3,5-triazine; 2-octylmercapto-4,6-bis-

(3,5-di-t-butyl-4-hydroxyphenoxy)-1,3,5-triazine; 2,4,6-tris-(3,5-di-t-butyl-4-hydroxyphenoxy)-1,2,3-triazine; 1,3,5-tris(3,5-di-t-butyl-4-hydroxybenzyl)isocyanurate; 1,3,5-tris(4-t-butyl-3-hydroxy-2,6-dimethylbenzyl)isocyanurate; 2,4,6-tris-(3,5-di-t-butyl-4-hydroxyphenylethyl)-1,3,5-triazine; 1,3,5-tris(3,5-di-t-butyl-4-hydroxyphenylpropionyl)hexahydro-1,3,5-triazine; 1,3,5-tris(3,5-dicyclohexyl-4-hydroxybenzyl)isocyanurate.

1.11 Benzylphosphonates such as, for example: dimethyl-2,5-di-t-butyl-4-hydroxybenzylphosphonate; diethyl-3,5-di-t-butyl-4-hydroxybenzylphosphonate; dioctadecyl-3,5-di-t-butyl-4-hydroxybenzylphosphonate; dioctadecyl-5-t-butyl-4-hydroxy-3-methylbenzylphosphonate; calcium salts of monoethyl ester of 3,5-di-t-butyl-4-hydroxybenzylphosphonic acid.

1.12 Acylaminophenols such as, for example: 4-hydroxylauranilide; 4-hydroxystearanilide; octyl-N-(3,5-di-t-butyl-4-hydroxyphenyl)carbamate.

1.13 Esters of  $\beta$ -(3,5-di-t-butyl-4-hydroxyphenyl)propionic acid with monohydric or polyhydric alcohols such as, for example: methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hy-

droxyethyl)oxalamide, 3-thioundecanol, 3-thiopentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospho-2,6,7-trioxabicyclo-[2.2.2]-octane.

1.14 Esters of  $\beta$ -(5-t-butyl-4-hydroxy-3-methylphenyl)propionic acid with monohydric or polyhydric alcohols such

5 as, for example: methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, 10 tris(hydroxyethyl)isocyanurate, N,N'-bis-(hydroxyethyl)oxamide, 3-thioundecanol, 3-thiopentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospho-2,6,7-trioxabicyclo[2.2.2]octane.

15 1.15 Esters of  $\beta$ -(3,5-dicyclohexyl-4-hydroxyphenyl)propionic acid with monohydric or polyhydric alcohols such

as, for example: methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, 20 tris(hydroxyethyl)isocyanurate, N,N'-bis-(hydroxyethyl)oxamide, 3-thioundecanol, 3-thiopentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospho-2,6,7-trioxabicyclo[2.2.2]-octane.

25 1.16 Esters of (3,5-di-t-butyl-4-hydroxyphenyl) acetic

acid with monohydric or polyhydric alcohols such as, for example: methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)-oxamide, 3-thioundecanol, 3-thiopentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospho-2,6,7-trioxabicyclo[2.2.2]octane.

10 1.17 Amides of  $\beta$ -(3,5-di-t-butyl-4-hydroxyphenyl)propionic acid such as, for example: N,N'-bis(3,5-di-t-butyl-4-hydroxyphenylpropionyl)hexamethylenediamide; N,N'-bis(3,5-di-t-butyl-4-hydroxyphenylpropionyl)trimethylenediamide; N,N'-bis(3,5-di-t-butyl-4-hydroxyphenylpropionyl)hydrazide; N,N'-bis[2-(3-[3,5-di-t-butyl-4-hydroxyphenyl]propionyloxy)ethyl]oxamide (Naugard<sup>®</sup> XL-1 of Uniroyal).

15 1.18 Ascorbic acid (vitamin C).

1.19 Aminic antioxidants such as, for example, N,N'-diisopropyl-p-phenylenediamine; N,N'-di-s-butyl-p-phenylenediamine; N,N'-bis(1,4-dimethylpentyl)-p-phenylenediamine; N,N'-bis(1-ethyl-3-methylpentyl)-p-phenylenediamine; N,N'-bis(1-methylheptyl)-p-phenylenediamine; N,N'-dicyclohexyl-p-phenylenediamine; N,N'-diphenyl-p-phenylenediamine; N,N'-bis(2-naphthyl)-p-phenylenediamine; N-isopropyl-N'-phenyl-p-phenylenediamine; N-(1,3-dimethyl-

20  
25

butyl)-N'-phenyl-p-phenylenediamine; N-(1-methylheptyl)-  
N'-phenyl-p-phenylenediamine; N-cyclohexyl-N'-phenyl-p-  
phenylenediamine; 4-(p-toluenesulfonamide)diphenylene-  
amine; N, N'-dimethyl-N,N'-di-s-butyl-p-phenylenediamine;  
5 diphenylamine; N-allyldiphenylamine; 4-isopropoxy-  
diphenylamine; N-phenyl-1-naphthylamine; N-(4-t-octyl-  
phenyl)1-naphthylamine; N-phenyl-2-naphthylamine; di-  
phenylamine octylate such as, for example, p,p'-di-t-  
octyldiphenylamine; 4-n-butylaminophenol; 4-butiryl-  
10 aminophenol; 4-nonanoylaminophenol; 4-dodecanoyl-  
aminophenol; 4-octadecanoylaminophenol; bis(4-  
methoxyphenyl)amine; 2,6-di-t-butyl-4-dimethylamino-  
methylphenol; 2,4'-diaminodiphenylmethane; 4,4'-diamino-  
diphenylmethane; N,N,N',N'-tetramethyl-4,4'-diaminodi-  
15 phenylmethane; 1,2-bis[(2-methylphenyl)amino] ethane;  
1,2-bis(phenylamino)propane; (o-tolyl)biguanide; bis[4-  
(1',3'-dimethylbutyl)phenyl]amine; N-phenyl-1-naphthyl-  
amine t-octylate; mixture of mono- and dialkylated t-  
butyl/t-octyldiphenylamines; mixture of mono- and dial-  
20 kylated nonyldiphenylamines; mixture of mono- and dial-  
kylated dodecyldiphenylamines; mixture of mono- and dial-  
kylated isopropyl/isohexyldiphenylamines; mixture of  
mono- and dialkylated t-butyldiphenylamines; 2,3-dihydro-  
3,3-dimethyl-4H-1,4-benzothiazine; phenothiazine; mixture  
25 of mono- and dialkylated t-butyl/t-octylphenothiazines;

mixture of mono- and dialkylated t-octyl-phenothiazines;  
N-allyl-phenothiazine; N,N,N',N'-tetraphenyl-1,4-diamino-  
but-2-ene; N,N-bis(2,2,6,6-tetramethylpiperid-4-yl)hexa-  
methylenediamine; bis-(2,2,6,6-tetramethylpiperidin-4-yl)  
5 sebacate; 2,2,6,6-tetramethylpiperidin-4-one; 2,2,6,6-  
tetramethylpiperidin-4-ol.

## 2. UV ray and light stabilizers.

### 2.1 Derivatives of 2-(2'-hydroxyphenyl)benzotriazoles

such as, for example: 2-(2'-hydroxy-5'-methylphenyl)-  
10 benzotriazole; 2-(3',5'-di-t-butyl-2'-hydroxyphenyl)ben-  
zotriazole; 2-(5'-t-butyl-2'-hydroxyphenyl)benzotriazole;  
2-[2'-hydroxy-5'-(1,1,3,3-tetramethylbutyl)phenyl]benzo-  
triazole; 2-(3',5'-di-t-butyl-2'-hydroxyphenyl)-5-chlo-  
robenzotriazole; 2-(3'-t-butyl-2'-hydroxy-5'-methylphe-  
15 nyl)-5-chlorobenzotriazole; 2-(3'-s-butyl-5'-t-butyl-2'-  
hydroxyphenyl)benzotriazole; 2-(2'-hydroxy-4'-octyloxy-  
phenyl)benzotriazole; 2-(3',5'-di-t-amyl-2'-hydroxyphe-  
nyl)benzotriazole; 2-[3',5'-bis( $\alpha,\alpha$ -dimethylbenzyl)-2'-  
hydroxyphenyl]benzotriazole; 2-[3'-t-butyl-2'-hydroxy-5'-  
20 (2-octyloxycarbonyl)ethyl]phenyl]-5-chlorobenzotriazole,  
2-[3'-t-butyl-5'-(2-(2-ethylhexyloxy)carbonyl)ethyl]-2'-  
hydroxyphenyl]-5-chlorobenzotriazole, 2-[3'-t-butyl-2'-  
hydroxy-5'-(2-methoxycarbonyl)ethyl]phenyl]-5-chloroben-  
zotriazole, 2-[3'-t-butyl-2'-hydroxy-5'-(2-methoxycarbo-  
25 nylethyl)phenyl]benzotriazole, 2-[3'-t-butyl-2'-hydroxy-



5'- (2-octyloxycarbonylethyl)phenyl]benzotriazole, 2-[3'-t-butyl-5'-(2-(2-ethylhexyloxy) carbonylethyl)-2'-hydroxyphenyl]benzotriazole, 2-(3'-dodecyl-2'-hydroxy-5'-methylphenyl)benzotriazole; 2-[3'-t-butyl-2'-hydroxy-5'-(2-iso-octyloxycarbonylethyl)phenyl]benzotriazole, 2,2'-methylen-bis[4-(1,1,3,3-tetramethylbutyl)-6-benzotriazol-2-yl-phenol]; transesterification product of 2-[3'-t-butyl-5'-(2-methoxycarbonylethyl)-2'-hydroxyphenyl]-2H-benzotriazole with polyethylene glycol 300;

10 [R-CH<sub>2</sub>CH<sub>2</sub>-COO-CH<sub>2</sub>CH<sub>2</sub>-]<sub>2</sub>-wherein R = 3'-t-butyl-4'-hydroxy-5'-2H-benzotriazol-2-yl-phenyl; 2-[2'-hydroxy-3'-( $\alpha,\alpha$ -dimethylbenzyl)-5'-(1,1,3,3-tetramethylbutyl)phenyl]benzotriazole; 2-[2'-hydroxy-3'-(1,1,3,3-tetramethylbutyl)-5'-( $\alpha,\alpha$ -dimethylbenzyl)phenyl]benzotriazole.

15 2.2 Benzotriazoles deriving from hydantoin such as those described, for example in patent applications EP 867,435, WO 99/23093 and WO 99/37638.

2.3 Derivatives of 2-hydroxybenzophenones such as, for example: 4-hydroxy-; 4-methoxy-; 4-octyloxy-; 4-decyloxy-; 20 ; 4-dodecyloxy-; 4-benzyloxy-; 4,2',4'-trihydroxy-; 2'-hydroxy-4,4'-dimethoxy.

2.4 Esters of benzoic acids, optionally substituted, such as, for example: phenyl salicylate, 4-t-butylphenyl salicylate, octylphenyl salicylate, benzoyl resorcinol, 25 bis(4-t-butylbenzoyl)resorcinol, dibenzoyl resorcinol,

2,4-di-t-butylphenyl-3,5-di-t-butyl-4-hydroxybenzoate,  
hexadecyl-3,5-di-t-butyl-4-hydroxybenzoate, octadecyl-  
3,5-di-t-butyl-4-hydroxybenzoate, 2-methyl-4,6-di-t-bu-  
tylphenyl-3,5-di-t-butyl-4-hydroxybenzoate.

5 2.5 Acrylates such as, for example, ethyl or iso-octyl  
 $\alpha$ -cyano- $\beta,\beta$ -diphenylacrylate; methyl  $\alpha$ -carbomethoxy-  
cinnamate, methyl or butyl  $\alpha$ -cyano- $\beta$ -methyl-p-methoxy-  
cinnamate, methyl  $\alpha$ -carbomethoxy-p-methoxycinnamate, N-  
( $\beta$ -carbomethoxy- $\beta$ -cyanovinyl)-2-methylindoline.

10 2.6 Nickel compounds such as, for example, Ni-complexes  
of 2,2'-thio-bis-[4-(1,1,3,3-tetramethylbutyl)phenol],  
for example 1:1 or 1:2 complexes, with or without addi-  
tional ligands such as n-butylamine, triethanolamine or  
N-cyclohexyldiethanolamine, nickel dibutyldithiocarbama-  
15 te, nickel salts of monoalkyl esters of 4-hydroxy-3,5-di-  
t-butyl-benzylphosphonic acid, such as methyl or ethyl  
esters, nickel complexes with ketoximes such as 2-  
hydroxy-4-methylphenyl undecyl ketoxime, nickel complexes  
of 1-phenyl-4-lauroyl-5-hydroxypyrazol with or without  
20 additional ligands.

2.7 Sterically hindered amines and their N-alkoxy de-  
rivatives such as, for example: poly-methylpropyl-3-oxy-  
[4-(2,2,6,6-tetramethyl)piperidinyl]siloxane, polymethyl-  
propyl-3-oxy-[4-(1,2,2,6,6-pentamethyl)piperidinyl]silo-  
25 xane, bis-(2,2,6,6-tetramethyl-4-piperidinyl)sebacate;

bis(2,2,6,6-tetramethyl-4-piperidiny) succinate; bis  
(1,2,2,6,6-pentamethyl-4-piperidiny) sebacate; bis(1-octyloxy-2,2,6,6-tetramethyl-4-piperidiny) sebacate; bis-  
(1,2,2,6,6-pentamethyl-4-piperidyl)-n-butyl-3,5-di-t-butyl-4-hydroxybenzylmalonate; condensation product between 1-(2-hydroxyethyl)-2,2,6,6-tetramethyl-4-hydroxy-piperidine and succinic acid; condensation product, linear or cyclic, between N,N'-bis(2,2,6,6-tetramethyl-4-piperid-yl) hexamethylenediamine and 4-t-octylamino-2,6-dichloro-1,3,5-s-triazine; tris(2,2,6,6-tetramethyl-4-piperidyl) nitrilotriacetate; tetrakis(2,2,6,6-tetramethyl-4-piperidyl)-1,2,3,4-butanetetracarboxylate; 1,1'-(1,2-ethanodiyl) bis(3,3,5,5-tetramethylpiperazinone; 4-benzoyl-2,2,6,6-tetramethylpiperidine; 4-stearyloxy-2,2,6,6-tetramethylpiperidine; bis(1,2,2,6,6-penta-methylpiperidyl)-2-n-butyl-2-(2-hydroxy-3,5-di-t-butylbenzyl)malonate; 3-n-octyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decane-2,4-dione; bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl) sebacate; bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl) succinate; condensation product, linear or cyclic, between N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl) hexamethylenediamine and 4-morpholine-2,6-di-chloro-1,3,5-triazine; condensation product between 2-chloro-4,6-di-(4-n-butylamino-2,2,6,6-tetramethyl-piperidyl)-1,3,5-triazine and 1,2-bis(3-aminopropylamino) ethane; condensation product be-

tween 2-chloro-4,6-di-(4-n-butylamino-1,2,2,6,6-pentamethylpiperidyl)-1,3,5-triazine and 1,2-bis(3-aminopropylamino)ethane; 8-acetyl-3-dodecyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro-[4.5]decane-2,4-dione; 3-dodecyl-1-  
5 (2,2,6,6-tetramethyl-4-piperidyl)pyrrolidin-2,5-dione; 3-dodecyl-1-(1,2,2,6,6-pentamethyl-4-piperidyl)pyrrolidin-2,5-dione; mixture of 4-hexadecyloxy- and 4-stearyloxy-2,2,6,6-tetramethylpiperidine; condensation product between N-N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-cyclohexylamino-2,6-dichloro-1,3,5-  
10 triazine; condensation product between 1,2-bis(3-aminopropylamino)ethane and 2,4,6-trichloro-1,3,5-triazine, as well as 4-butylamino-2,2,6,6-tetramethylpiperidine (CAS Reg. Nr. [136504-96-6]; N-(2,2,6,6-tetramethyl-4-piperidyl)-n-dodecylsuccinimide; N-(1,2,2,6,6-pentamethyl-4-piperidyl)-n-dodecylsuccinimide; 2-undecyl-7,7,9,9-tetramethyl-1-oxa-3,8-diaza-4-oxospiro[4,5]decane; reaction product between 7,7,9,9-tetramethyl-2-cycloundecyl-1-oxa-3,8-diaza-4-oxospiro-[4,5]decane and epichlorohydrin;  
15 1,1-bis(1,2,2,6,6-pentamethyl-4-piperidyloxycarbonyl)-2-(4-methoxyphenyl)ethene; N,N'-bis-formyl-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine; diester of 4-methoxy-methylenemalononic acid with 1,2,2,6,6-pentamethyl-4-hydroxypiperidine; reaction product of maleic  
20 anhydride/ $\alpha$ -olefin copolymer with 2,2,6,6-tetramethyl-4-

aminopiperidine or with 1,1,2,2,6-pentamethyl-4-aminopiperidine.

2.8 Oxamides such as, for example: 4,4'-dioctyloxyoxanilide; 2,2'-diethoxyoxanilide; 2,2'-dioctyloxy-5,5'-di-t-butoxanilide; 2,2'-didodecyloxy-5,5'-di-t-butyloxanilide; 2-ethoxy-2'-ethyloxanilide; N,N'-bis(3-dimethylaminopropyl)oxamide; 2-ethoxy-5-t-butyl-2'-ethyloxanilide and its mixtures with 2-ethoxy-2'-ethyl-5,4'-di-t-butoxanilide; and mixtures of di-substituted ortho- and para-methoxy oxanilides and mixtures of di-substituted ortho and para-ethoxy oxanilides.

2.9 2-(2-hydroxyphenyl)-1,3,5-triazines such as, for example: 2,4,6-tris(2-hydroxy-4-octyloxyphenyl)-1,3,5-triazine; 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-(2,4-dihydroxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2,4-bis-(2-hydroxy-4-propyloxyphenyl)-6-(2,4-dimethylphenyl)-1,3,5-triazine; 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(4-methylphenyl)-1,3,5-triazine; 2-(2-hydroxy-4-dodecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-[2-hydroxy-4-(2-hydroxy-3-butyloxypropyloxy)phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-[2-hydroxy-4-(2-hydroxy-3-octyloxypropyloxy)phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-(2-hydroxy-4-tridecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-

[4-(dodecyloxy/tridecyloxy-2-hydroxypropoxy)-2-hydroxy-phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-[2-hydroxy-4-(2-hydroxy-3-dodecyloxypropoxy)phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-(2-hydroxy-4-hexyloxyphenyl)-4,6-diphenyl-1,3,5-triazine; 2-(2-hydroxy-4-methoxyphenyl)-4,6-diphenyl-1,3,5-triazine; 2,4,6-tris[2-hydroxy-4-(3-butoxy-2-hydroxypropoxy)phenyl]-1,3,5-triazine; 2-(2-hydroxyphenyl)-4-(4-methoxyphenyl)-6-phenyl-1,3,5-triazine; 2-{2-hydroxy-4-[3-(2-ethylhexyl-1-oxy)-2-hydroxypropyloxy]phenyl}-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine.

3. "Metal-deactivators" such as, for example: N,N'-diphenyloxamide, N-salicylal-N'-salicyloyl-hydrazine, N,N'-bis(salicyloyl)hydrazine, N,N'-bis(3,5-di-t-butyl-4-hydroxyphenylpropionyl)hydrazine, 3-salicyloylamino-1,2,4-triazole, bis(benzylidene)oxallyl dihydrazide, oxanilide, isophthaloyl dihydrazide, sebacoyl bisphenylhydrazide, N,N'-diacetyladipoyl dihydrazide, N,N'-bis(salicyloyl)oxallyl dihydrazide, N,N'-bis(salicyloyl)thiopropionyl dihydrazide.

4. Phosphites and phosphonites such as, for example: triphenyl phosphite, diphenyl alkyl phosphites, phenyl dialkyl phosphites, tris(nonylphenyl)phosphite, trilauryl phosphite, trioctadecyl phosphite, distearyl pentaerythritol diphosphite, tris(2,4-di-t-butylphenyl) phosphite,

diisodecyl pentaerythritol diphosphite, bis(2,4-di-t-butylphenyl) pentaerythritol diphosphite, bis(2,6-di-t-butyl-4-methylphenyl)pentaerythritol diphosphite, diisodecyloxypentaerythritol diphosphite, bis(2,4-di-t-butyl-6-methylphenyl)pentaerythritol diphosphite, bis [2,4,6-tris(t-butylphenyl)]pentaerythritol diphosphite, tristearyl sorbitol triphosphite, tetrakis-(2,4-di-t-butylphenyl)-4,4'-diphenylenediphosphonite, 6-isooctyloxy-2,4,8,10-tetra-t-butyl-12H-dibenzo-[d,g]-1,3,2-dioxaphosphocine, 6-fluoro-2,4,8,10-tetra-t-butyl-12-methyldibenzo[d,g]-1,3,2-dioxaphosphocine, bis-(2,4-di-t-butyl-6-methylphenyl)methylphosphite, bis(2,4-di-t-butyl-6-methylphenyl)ethylphosphite; 2,2',2''-nitrilo [triethyl-tris (3,3',5,5'-tetra-t-butyl-1,1'-biphenyl-2,2'-diyl)-phosphite]; 2-ethylhexyl-(3,3',5,5'-tetra-t-butyl-1,1'-biphenyl-2,2'-diyl)phosphite.

5. Hydroxylamines such as, for example: N,N-dibenzylhydroxylamine; N,N-diethylhydroxylamine; N,N-dioctylhydroxylamine; N,N-dilaurylhydroxylamine; N,N-ditetradecylhydroxylamine; N,N-dihexadecylhydroxylamine; N,N-dioctadecylhydroxylamine; N-hexadecyl-N-octadecylhydroxylamine; N-heptadecyl-N-octadecylhydroxylamine; N,N-dialkylhydroxylamines deriving from hydrogenated tallow amines.

6. Nitrons such as, for example: N-benzyl- $\alpha$ -phenyl-

nitron; N-ethyl- $\alpha$ -methyl-nitron; N-octyl- $\alpha$ -heptyl-nitron;  
N-lauryl- $\alpha$ -undecyl-nitron; N-tetradecyl- $\alpha$ -tridecyl-ni-  
tron; N-hexadecyl- $\alpha$ -pentadecyl-nitron; N-octadecyl- $\alpha$ -  
heptadecyl-nitron; N-hexadecyl- $\alpha$ -heptadecyl-nitron; N-  
5 octadecyl- $\alpha$ -pentadecyl-nitron; N-heptadecyl- $\alpha$ -heptadecyl-  
nitron; N-octadecyl- $\alpha$ -hexadecyl-nitron; nitrons deriving  
from hydrogenated tallow amines.

7. Thiosynergizing agents such as, for example: dilauryl  
thiodipropionate; distearyl thiodipropionate.

10 8. Agents which are capable of destroying peroxides such  
as, for example, esters of  $\beta$ -thiodipropionic acid such as  
lauryl, stearyl, myristyl or tridecyl esters, mercapto-  
benzimidazole or zinc salt of 2-mercaptobenzimidazole,  
zinc dibutyldithiocarbamate, dioctadecyldisulfide, pen-  
15 taerythritol tetrakis( $\beta$ -dodecylmercapto)propionate.

9. Polyamide stabilizers such as, for example, copper  
salts combined with compounds of iodine and/or phospho-  
rous, divalent manganese salts.

10. Basic co-stabilizers such as, for example: melamine,  
20 polyvinylpyrrolidone, dicyanodiamide, triallyl cyanurate,  
derivatives of urea, derivatives of hydrazine, amines,  
polyamides, polyurethanes, salts of alkaline metals and  
salts of earth-alkaline metals of fatty acids with a high  
molecular weight such as, for example, Ca-stearate, Zn-  
25 stearate, Mg-stearate, Mg-behenate, Na-ricinoleate, K-



palmitate, antimonium-pyrocatecholate, tin-pyrocatecholate, zinc-pyrocatecholate.

11. Nucleating agents such as, for example: inorganic substances such as talc, metal oxides (for example, titanium dioxide or magnesium oxide), phosphates, carbonates or sulfates (preferably of earth-alkaline metals); organic compounds such as mono- or polycarboxylic acids and their salts (for example, 4-t-butylbenzoic acid, adipic acid, diphenylacetic acid, sodium succinate, sodium benzoate); polymeric compounds such as ionic copolymers ("ionomers").

12. Fillers and reinforcing agents such as, for example: calcium carbonate, silicates, glass fibres, glass beads, asbestos, talc, kaolin, mica, barium sulfate, metal oxides and hydroxides, carbon black, graphite, wood flour and flours or fibres of other natural products, synthetic fibres.

13. Other additives such as, for example: plasticizers, pigments, lubricants, emulsifying agents, rheological additives, catalysts, slip agents, optical brighteners, flame-retardants (for example bromurates, chlorurates, phosphorates and phosphorous/halogen mixtures), anti-static agents, blowing agents.

14. Benzofuranones and indolinones such as, for example: 3-[4-(2-acetoxyethoxy)phenyl]-5,7-di-t-butyl-benzofuran-

2-one; 5,7-di-t-butyl-3-[4-(2-stearoyloxyethoxy)phenyl]-  
benzofuran-2-one; 3,3'-bis[5,7-di-t-butyl-3-[4-(2-hyd-  
roxyethoxy)phenyl]benzofuran-2-one; 5,7-di-t-butyl-3-(4-  
ethoxyphenyl)benzofuran-2-one; 3-(4-acetoxy-3,5-dimethyl-  
5 phenyl)-5,7-di-t-butyl-benzofuran-2-one; 3-(3,5-dimethyl-  
4-pivaloyloxyphenyl)-5,7-di-t-butyl-benzofuran-2-one; or  
those described in U.S. patents Nr. 4,325,863, 4,338,244,  
5,175,312, 5,216,052 and 5,252,643; in German patents DE  
4,316,611, 4,316,622 and 4,316,876; or in European patent  
10 applications Nr. 589,839 and 591,102.

The above stabilizers (co-stabilizers) can be option-  
ally added to the organic polymers to be stabilized in a  
quantity ranging from 0.01% to 10% with respect to the to-  
tal weight of the organic polymer to be stabilized.

15 The incorporation of compounds (a), (b), (c) and, op-  
tionally, (d), as single compounds or mixed with each  
other and possibly other stabilizers (co-stabilizers), in  
the organic polymers to be stabilized, can be carried out  
according to the methods known in the art, for example,  
20 before or during the processing, or the mixture dissolved  
or dispersed in a solvent, before or after its evapora-  
tion, can be applied to the organic polymer to be stabi-  
lized. The stabilizing mixture, object of the present in-  
vention, can also be used in masterbatch form containing  
25 from 2.5% to 25% by weight of said mixture.

The above mixture, optionally in the presence of other stabilizers (co-stabilizers), can also be added to the organic polymers to be stabilized, either after or during polymerization or before cross-linking.

5       The above mixture, optionally in the presence of other stabilizers (co-stabilizers), can be added to the organic polymers to be stabilized, in pure form or encapsulated in waxes, oils or polymers.

10       The above mixture, optionally in the presence of other stabilizers (co-stabilizers) which can be diluted or used in the molten state, can be sprayed into the organic polymers to be stabilized. This spraying can be advantageously effected during the deactivation of the polymerization catalyst, as the spraying can be effected using, 15 for example, the vapor used for the deactivation.

In the case of spherically polymerized polyolefins, it may be advantageous to add the above mixture, optionally in the presence of other stabilizers (co-stabilizers), by means of spraying.

20       The organic polymers stabilized as described above can be used in a wide variety of forms such as, for example, films, fibers, tapes, moulding compositions, profiles, ligands for coating materials such as powder coatings, adhesives or plaster, in particular.

25       The present invention also relates to a method for

stabilizing organic polymers against degradation caused by oxygen, heat and/or light, which comprises the addition or application to said organic polymers of the stabilizing mixture, object of the present invention.

5       The 3-pyrazolidinones having general formula (I) or (II) can be used as such, as process stabilizers for organic polymers. A further object of the present invention is consequently the use of said compounds as process stabilizers for organic polymers, the polymeric compositions  
10   thus stabilized and the end-products obtained from their processing.

Some illustrative but non-limiting examples are provided hereunder for a better understanding of the present invention and for its embodiment.

15   EXAMPLE 1

**Stabilization of polypropylene subjected to multiple high temperature extrusions**

100 g of polypropylene in powder (Moplen FLF 20 of Montell Italia), having a melt flow index (MFI) of 12  
20   g/min measured in accordance with ASTM D638 at 230°C with 2.16 kg, are mixed with 0.05 g of calcium stearate and with the other stabilizers indicated in Table 1, which also specifies the quantities of the stabilizers used.

The above homogenized mixture is fed to a Brabender  
25   laboratory extruder with a feeding screw having a length

of 475 mm, a diameter of 19 mm, a compression ratio of 1:4, which has the possibility of differentiated heating in four different zones of the feeding screw. The mixture is extruded through a hole having a diameter of 2 mm, with  
 5 a screw rotation rate of 60 rpm and a temperature profile of 190°C, 230°C, 250°C, 280°C and the MFI values obtained, measured as described above, at the 1<sup>st</sup>, 3<sup>rd</sup> and 5<sup>th</sup> extrusion, are indicated in Table 1.

Table 1

STABILIZING MIXTURE	MFI		
	1 <sup>st</sup>	3 <sup>rd</sup>	5 <sup>th</sup>
Anox 20 (0.0375%) Alkanox 240 (0.0375%)	14.8	18.7	24.3
Anox 20 (0.025%) Alkanox 240 (0.0025%) Compound (IA) (0.01%)	12.8	14.2	17.8
15 Anox 20 (0.02%) Alkanox 240 (0.02%) Compound (IA) (0.01%)	12.5	14.5	19.2
Anox 20 (0.02%) Alkanox 240 (0.02%) Compound (IA) (0.0075%)	13.7	16.2	20.7

20 EXAMPLE 2

**Stabilization of polypropylene subjected to multiple high temperature extrusions**

100 g of polypropylene in powder (Moplen FLF 20 of Montell Italia), having a melt flow index (MFI) of 18  
 25 g/min measured in accordance with ASTM D638 on a Ceast

Automatic Melt Flow Tester at 230°C and under a 2.16 kg load, are mixed with 0.05 g of calcium stearate and with the other stabilizers indicated in Table 2, which also specifies the quantities of the stabilizers used.

5        The above homogenized mixture is fed to a Brabender PL 200 single screw laboratory extruder with a feeding screw having a length of 475 mm, a diameter of 19 mm, a compression ratio of 1:4, which has the possibility of differentiated heating in four different zones of the  
10        feeding screw. The mixture is extruded through a hole having a diameter of 2 mm, with a screw rotation rate of 60 rpm and a temperature profile of 200°C, 225°C, 250°C, 275°C and the MFI values obtained, measured as described above, at the 1<sup>st</sup>, 3<sup>rd</sup> and 5<sup>th</sup> extrusion, are indicated in  
15        Table 2.

Table 2

STABILIZING MIXTURE	MFI		
	1 <sup>st</sup>	3 <sup>rd</sup>	5 <sup>th</sup>
Alkanox 240 (0.025%) LL 62 (0.025%) Compound (IA) (0.025%)	11.2	11.9	13.6
Alkanox 240 (0.025%) LL 62 (0.025%) *Compound (IA) (0.025%)	11.4	11.9	13.2
Alkanox 240 (0.025%) LL 62 (0.025%) **Compound (IA) (0.025%)	11.3	11.9	12.9

\*Compound (IA) was crystallized after treatment with active charcoal;

\*\* Compound (IA) was crystallized twice from methylene Chloride/Cyclohexane 1:3 v/v.

- 5 The Yellow Index (YI) was measured on the granules after each extrusion for the polypropylene formulations reported in Table 2 using a Macbeth Colour Eye instrument mounting a light source D65 according to ASTM-E-313 and the data are reported in Table 3

10 Table 3

STABILIZING MIXTURE	YI		
	1 <sup>st</sup>	3 <sup>rd</sup>	5 <sup>th</sup>
Alkanox 240 (0.025%) LL 62 (0.025%) Compound (IA) (0.025%)	-0.17	2.59	5.62
Alkanox 240 (0.025%) LL 62 (0.025%) *Compound (IA) (0.025%)	-0.58	2.40	5.36
Alkanox 240 (0.025%) LL 62 (0.025%) **Compound (IA) (0.025%)	-0.82	1.40	3.91

\*Compound (IA) was crystallized after treatment with active charcoal;

- 15 \*\* Compound (IA) was crystallized twice from methylene Chloride/Cyclohexane 1:3 v/v.

### EXAMPLE 3

Stabilization of polypropylene subjected to multiple high

**temperature extrusions**

100 g of polypropylene in powder form (Moplen FLF 20 of Montell Italia), having a melt flow index (MFI) equal to 18 g/min measured in accordance with the regulation 5 ASTM D638 on a Ceast Automatic Melt Flow Tester at 230°C and under a 2.16 kg load, are mixed with 0.05 g of calcium stearate and with the other stabilizers indicated in Table 4: Table 4 also specifies the quantities of the stabilizers used. The above homogenized mixture is fed to a Bra- 10 bender PL 200 single screw laboratory extruder with a feeding screw having a length of 475 mm, a diameter of 19 mm, a compression ratio of 1:4, which has the possibility of differentiated heating in four different zones of the feeding screw. The mixture is extruded through a hole hav- 15 ing a diameter of 2 mm, with a screw rotation rate of 60 rpm and a temperature profile of 200°C, 225°C, 250°C, 275°C and the MFI values obtained, measured as described above, at the 1<sup>st</sup>, 3<sup>rd</sup> and 5<sup>th</sup> extrusion, are indicated in Table 4.

20 Table 4

STABILIZING MIXTURE	MFI		
	1 <sup>st</sup>	3 <sup>rd</sup>	5 <sup>th</sup>
Alkanox 240 (0.075%)	15.3	22.1	30.8
Compound (IA) (0.05%)	11.9	13.2	15.9
Alkanox 240 (0.05%)	18.2	27.6	36.4
Compound (IA) (0.025%)	12.0	14.1	18.4



CLAIMS

1. Stabilizing mixtures for organic polymers comprising:

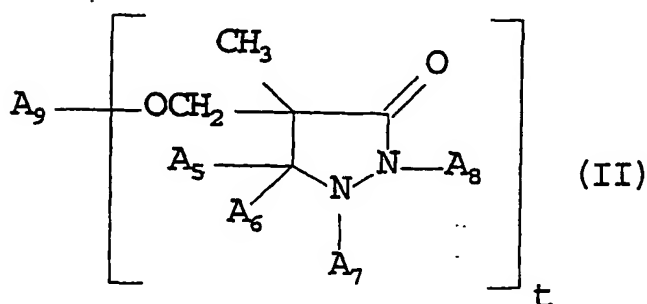
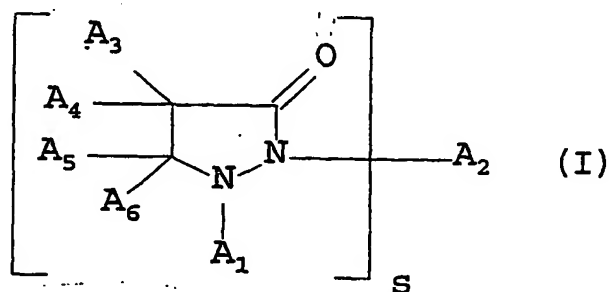
(a) at least one compound belonging to the group of  
5 3-pyrazolidinones;

(b) at least one compound belonging to the group or  
organic phosphites or phosphonites; and optionally

(c) at least one group belonging to the group of  
sterically hindered phenols; and, optionally,

10 (d) at least one group belonging to the group of  
sterically hindered amines.

2. The stabilizing mixtures for organic polymers ac-  
cording to claim 1, wherein the compounds belonging  
to the group of 3-pyrazolidinones (a) are selected  
15 from those having general formula (I) or (II):



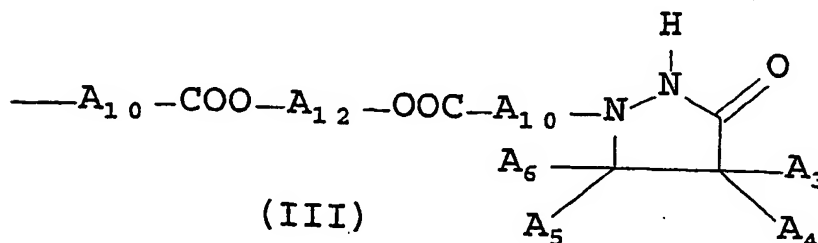
wherein:

- 5                   -    A<sub>1</sub> represents a linear or branched C<sub>1</sub>-C<sub>18</sub> alkyl group; a C<sub>5</sub>-C<sub>12</sub> cycloalkyl group, said cycloalkyl group optionally mono-, di- or tri-substituted with a linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl group; a phenyl group, said phenyl group optionally mono-, di- or tri-substituted with a linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl or alkoxy group, and/or mono-substituted with a hydroxyl group; a C<sub>7</sub>-C<sub>9</sub> phenylalkyl group, said phenylalkyl group optionally mono-, di- or tri-substituted on the phenyl with a linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl group, and/or mono-substituted with a hydroxyl group; an
- 10                   -A<sub>10</sub>-COOA<sub>11</sub> wherein A<sub>10</sub> represents a linear or branched C<sub>1</sub>-C<sub>6</sub> alkylene group and A<sub>11</sub> represents a linear or branched C<sub>1</sub>-C<sub>18</sub> alkyl group, a C<sub>5</sub>-C<sub>12</sub> cycloalkyl group, said cycloalkyl group optionally mono-, di- or tri-substituted with a linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl group, a linear or branched C<sub>3</sub>-C<sub>18</sub> alkenyl group, a C<sub>7</sub>-C<sub>9</sub> phenylalkyl group, said phenylalkyl group optionally mono-, di- or tri-substituted on the phenyl with a linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl group;
- 20                   -    A<sub>3</sub>, A<sub>4</sub>, A<sub>5</sub> and A<sub>6</sub>, the same or different, repre-
- 25

sent a hydrogen atom; a linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl group; a phenyl group;

- s is 1 or 2;

- when s is 1, A<sub>2</sub> represents a hydrogen atom; a linear or branched C<sub>1</sub>-C<sub>18</sub> alkyl group; a linear or branched C<sub>3</sub>-C<sub>6</sub> alkenyl group; a C<sub>7</sub>-C<sub>9</sub> phenylalkyl group, said phenylalkyl group optionally mono-, di- or tri-substituted on the phenyl with a linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl group; an -A<sub>10</sub>-COOA<sub>11</sub> wherein A<sub>10</sub> and A<sub>11</sub> have the same meanings defined above; and, when A<sub>2</sub> represents a hydrogen atom, A<sub>1</sub> may also represent a group having general formula (III):



- wherein A<sub>3</sub>, A<sub>4</sub>, A<sub>5</sub>, A<sub>6</sub> and A<sub>10</sub> have the same meanings defined above and A<sub>12</sub> represents a linear or branched C<sub>2</sub>-C<sub>12</sub> alkylene group; a C<sub>4</sub>-C<sub>12</sub> alkylene group interrupted by 1, 2 or 3 oxygen atoms; a cyclohexylene group; a cyclohexylenedimethylene group; an isopropylidenedi-cyclohexylidene group;

- when s is 2, A<sub>2</sub> represents a linear or branched

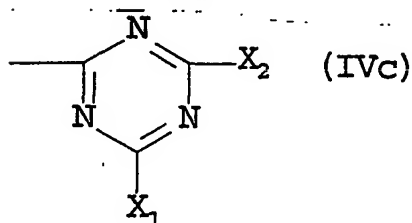
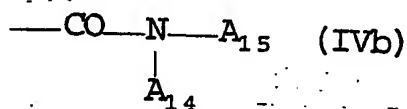
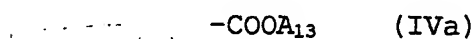
C<sub>2</sub>-C<sub>12</sub> alkylene group, or a xylylene group;

- A<sub>7</sub> represents a phenyl group optionally mono-, di- or tri-substituted with a linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl or alkoxy group, and/or mono-substituted with a hydroxyl group;

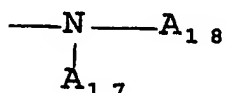
- A<sub>8</sub> represents a hydrogen atom; a linear or branched C<sub>1</sub>-C<sub>18</sub> alkyl group; a linear or branched C<sub>3</sub>-C<sub>6</sub> alkenyl group; a C<sub>7</sub>-C<sub>9</sub> phenylalkyl group, said phenylalkyl group optionally mono-, di-, or tri-substituted on the phenyl with a linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl group; an -A<sub>10</sub>-COOA<sub>11</sub> group wherein A<sub>10</sub> and A<sub>11</sub> have the same meanings defined above;

- t is 1, 2, 3 or 4;

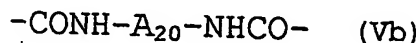
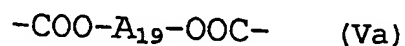
- when t is 1, A<sub>9</sub> represents an acyl-aliphatic, cycloaliphatic, aromatic, arylaliphatic or heterocyclic group containing not more than 22 carbon atoms; or one of the following groups having general formula (IVa)-(IVc):

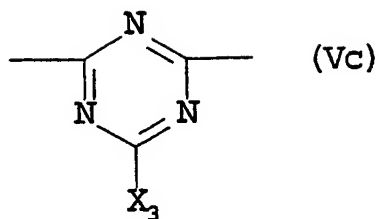


wherein  $A_{13}$  has the same meanings as  $A_{11}$  described above;  $A_{14}$  and  $A_{15}$ , the same or different, have the same meanings as  $A_{11}$  described above, or they represent a hydrogen atom, or a phenyl group, or  $A_{14}$  and  $A_{15}$  considered jointly with the nitrogen atom to which they are bound, represent a heterocyclic group with 5-7 atoms;  $X_1$  and  $X_2$ , the same or different, represent an -O- $A_{16}$  group, or a group having the formula:



wherein  $A_{16}$ ,  $A_{17}$  and  $A_{18}$ , the same or different, have the same meanings as  $A_{14}$  and  $A_{15}$  described above, or  $A_{17}$  and  $A_{18}$  considered jointly with the nitrogen atom to which they are bound, represent a heterocyclic group with 5-7 atoms; when  $t$  is 2,  $A_9$  represents a diacyl-aliphatic, cycloaliphatic, aromatic, arylaliphatic or heterocyclic group containing not more than 22 carbon atoms; or one of the following groups having general formula (Va)-(Vc):





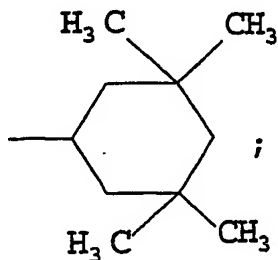
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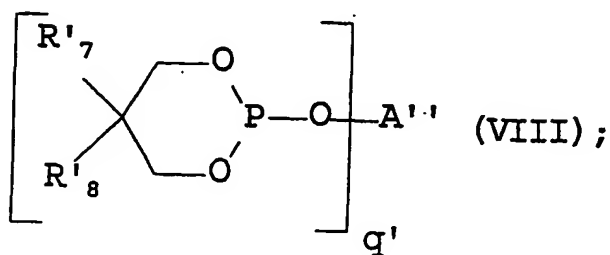
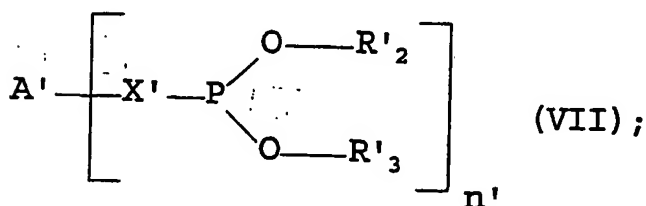
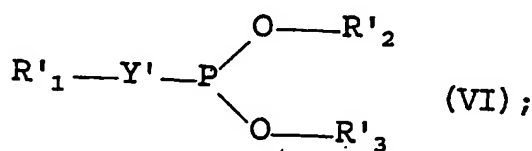
wherein  $A_{19}$  represents a linear or branched  $C_2$ - $C_{12}$  alkylene group, a  $C_4$ - $C_{12}$  alkylene group interrupted by 1, 2 or 3 oxygen atoms, a cyclohexylene group, a cyclohexylenedimethylene group, an isopropylidenedicyclohexylidene group, a phenylene group, a xylylene group, an isopropylidenediphenylene group;  $A_{20}$  represents a linear or branched  $C_2$ - $C_{12}$  alkylene group, a cyclohexylene group, a cyclohexylenedimethylene group, a methylenedicyclohexylene group, a phenylene group, a methylphenylene group, a xylylene group, a methylenediphenylene group, an oxydiphenylene group, a group having the formula:



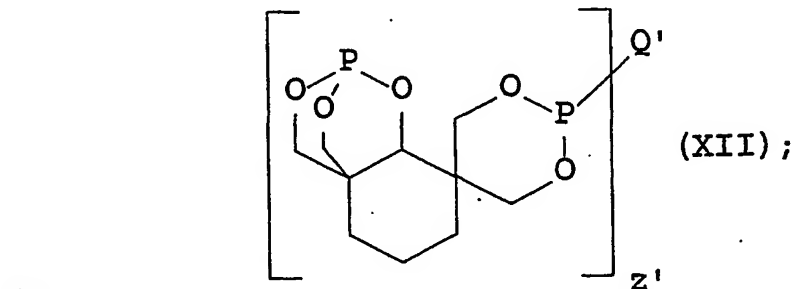
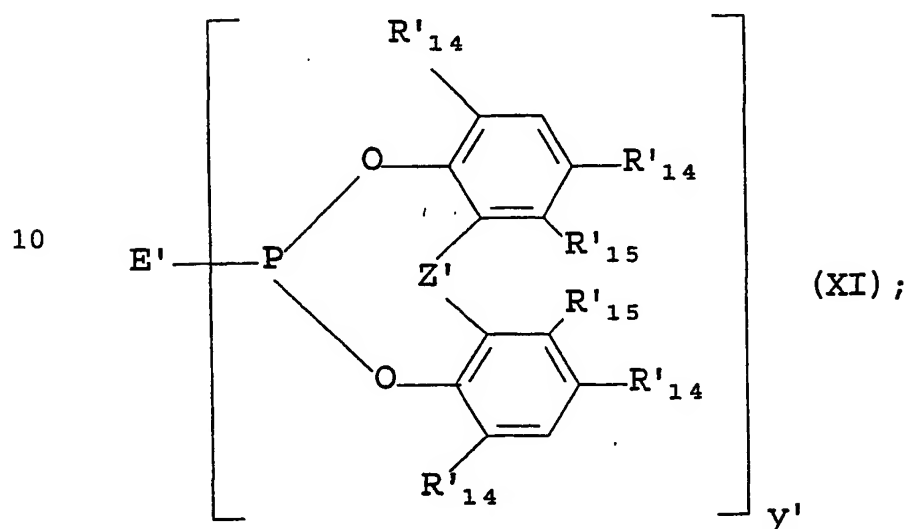
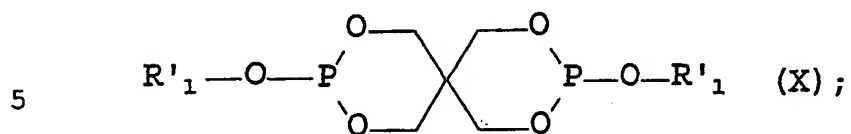
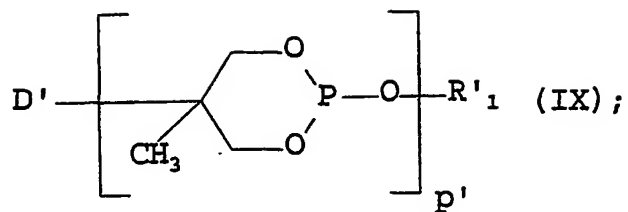
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$X_3$  has the same meanings as  $X_1$  and  $X_2$  defined above;

- when t is 3, A<sub>9</sub> represents a triacyl-aliphatic, aromatic or heterocyclic group containing not more than 18 carbon atoms, or a 2,4,6-triyl-1,3,5-triazine group;
- 5 - when n is 4, A<sub>9</sub> represents a tetra-acyl-aliphatic or aromatic group containing not more than 18 carbon atoms.
3. The stabilizing mixtures for organic polymers according to claim 1, wherein the compounds belonging to the group of organic phosphites or phosphonites
- 10 (b) are selected from those having the following general formulae (VI)-(XII):



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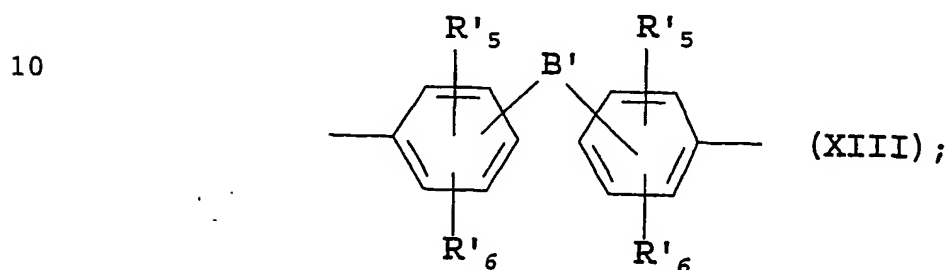
wherein:

- $n'$  is 2, 3 or 4;
- $p'$  is 1 or 2;
- $q'$  is 2 or 3;
- $r'$  is an integer ranging from 4 to 12, extremes

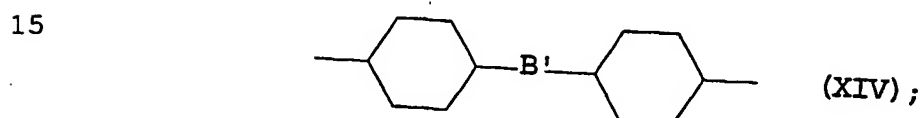


included;

- y' is 1, 2 or 3;
- z' is an integer ranging from 1 to 6, extremes included;
- 5 - when n' is 2, A' represents a C<sub>2</sub>-C<sub>18</sub> alkylene group; a C<sub>2</sub>-C<sub>12</sub> alkylene group containing an oxygen atom, a sulfur atom, or an -NR'<sub>4</sub>- group; a group having general formula (XIII):

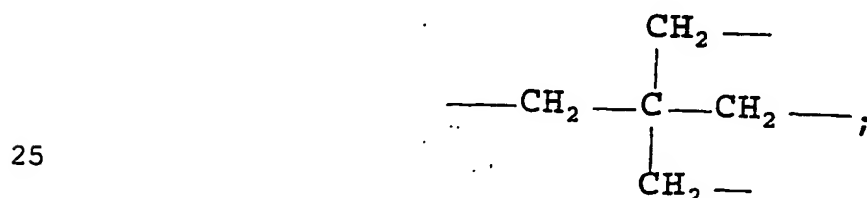


a group having general formula (XIV):



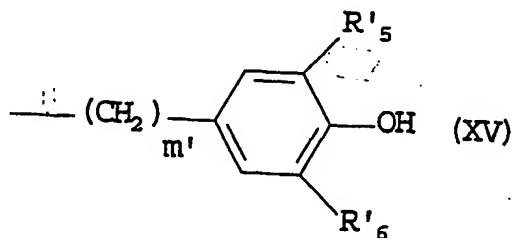
a phenylenic group;

- when n' is 3, A' represents a group having general formula -C<sub>r</sub>H<sub>2r'-1</sub>- wherein r' has the same
- 20 values described above;
- when n' is 4, A' represents a group having the formula:



- A' has the same meanings defined above for A' when n' is 2;
- B' represents a direct bond; one of the following groups:  $-\text{CH}_2-$ ,  $-\text{CHR}'_4-$ ,  $-\text{CR}'_1\text{R}'_4-$ ; a sulfur atom; a  $\text{C}_5$ - $\text{C}_7$  cycloalkylidene group; a cyclohexylidene group substituted with 1-4  $\text{C}_1$ - $\text{C}_4$  alkyl groups in position 3, 4 and/or 5;
- when p' is 1, D' represents a methyl group; and, when p' is 2, D' represents a  $-\text{CH}_2\text{OCH}_2-$  group;
- when y' is 1, E' represents a  $\text{C}_1$ - $\text{C}_{18}$  alkyl group; an  $-\text{OR}'_1$  group; a halogen atom;
- when y' is 2, E' represents an  $-\text{O}-\text{A}'-\text{O}-$  group;
- when y' is 3, E' represents one of the following groups:  $\text{R}'_4\text{C}(\text{CH}_2\text{O}-)_3$ ,  $\text{N}(\text{CH}_2\text{CH}_2\text{O}-)_3$ ;
- Q' represents a radical of an alcohol or a phenol with a valence z', said radical being attached to the phosphorous atom by means of an oxygen atom;
- $\text{R}'_1$ ,  $\text{R}'_2$  and  $\text{R}'_3$ , each independently, represent a hydrogen atom; a halogen atom; one of the following groups:  $-\text{COOR}'_4-$ ,  $-\text{CN}-$ ,  $-\text{CONR}'_4\text{R}'_4$ ; a  $\text{C}_2$ - $\text{C}_{18}$  alkyl group containing an oxygen atom, a sulfur atom, an  $-\text{NR}'_4-$  group; a  $\text{C}_7$ - $\text{C}_9$  phenylalkyl group; a  $\text{C}_5$ - $\text{C}_{12}$  cycloalkyl group; a phenyl

group or a naphthyl group, said phenyl or naphthyl groups optionally substituted with halogen atoms, or with 1-3 C<sub>1</sub>-C<sub>18</sub> alkyl or alkoxy groups or with C<sub>7</sub>-C<sub>9</sub> phenylalkyl groups; or they represent a group having general formula (XV):



wherein m' represents an integer ranging from 3 to 6, extremes included;

- R'<sub>4</sub> represents a hydrogen atom; a C<sub>1</sub>-C<sub>18</sub> alkyl group; a C<sub>5</sub>-C<sub>12</sub> cycloalkyl group; a C<sub>7</sub>-C<sub>9</sub> phenylalkyl group;
- R'<sub>5</sub> and R'<sub>6</sub> each independently, represent a hydrogen atom; a C<sub>1</sub>-C<sub>8</sub> alkyl group; a C<sub>5</sub>-C<sub>6</sub> cycloalkyl group;
- when q' is 2, R'<sub>7</sub> and R'<sub>8</sub> each independently, represent a C<sub>1</sub>-C<sub>4</sub> alkyl group; or, considered jointly, they represent a 2,3-dihydropentamethylene group;
- when q' is 3, R'<sub>7</sub> and R'<sub>8</sub> represent a methyl group;
- R'<sub>14</sub> represents a hydrogen atom; a C<sub>1</sub>-C<sub>9</sub> alkyl

group; a cyclohexyl group;

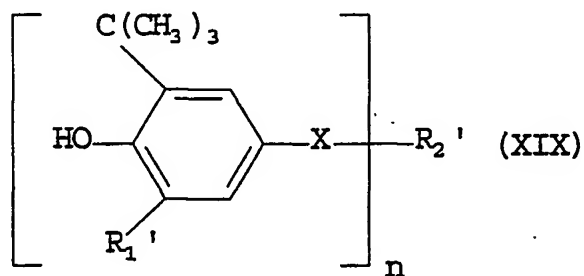
- R'<sub>15</sub> represents a hydrogen atom; a methyl group; or, when two or more R'<sub>14</sub> and R'<sub>15</sub> groups are present, said groups are the same or different to each other;

- X' and Y' represent a direct bond; an oxygen atom;

- Z' represents a direct bond; a methylene group; a -C(R'<sub>16</sub>)<sub>2</sub>- group; a sulfur atom;

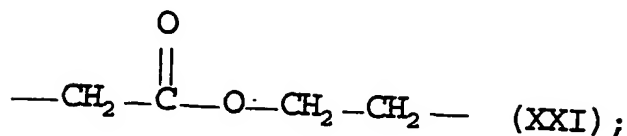
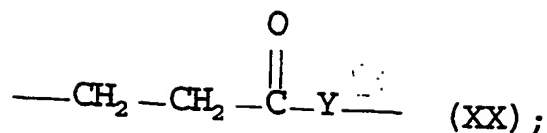
- R'<sub>16</sub> represents a C<sub>1</sub>-C<sub>8</sub> alkyl group.

4. The stabilizing mixtures for organic polymers according to claim 1, wherein the compounds belonging to the group of sterically hindered phenols (c) are selected from those having general formula (XIX):

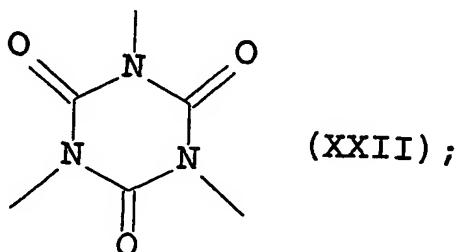


wherein:

- R'<sub>1</sub> represents a C<sub>1</sub>-C<sub>4</sub> alkyl group;
- n is 1, 2, 3 or 4;
- X represents a methylene group; or a group having general formula (XX) or (XXI):

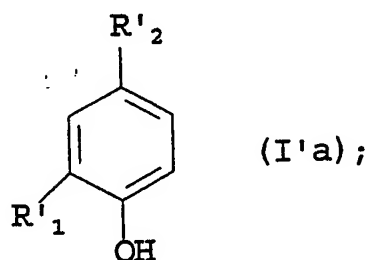
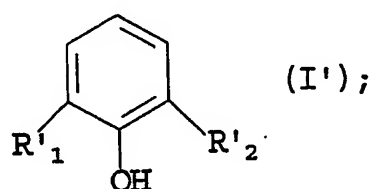


- 5        -    Y represents an oxygen atom; an -NH- group;
- when n is 1, X represents a group having general formula (XX) wherein Y is attached to R<sub>2</sub>' and R<sub>2</sub>' represents a C<sub>1</sub>-C<sub>25</sub> alkyl group;
- 10       -    when n is 2, X represents a group having general formula (XX) wherein Y is attached to R<sub>2</sub>' and R<sub>2</sub>' represents a C<sub>2</sub>-C<sub>12</sub> alkylene group; a C<sub>4</sub>-C<sub>12</sub> alkylene group containing one or more oxygen or sulfur atoms; or, when Y represents an -NH- group, R<sub>2</sub>' represents a direct bond;
- 15       -    when n is 3, X represents a methylene group; a group having general formula (XXI) wherein the ethylene group is bound to R<sub>2</sub>' and R<sub>2</sub>' represents a group having formula (XXII):

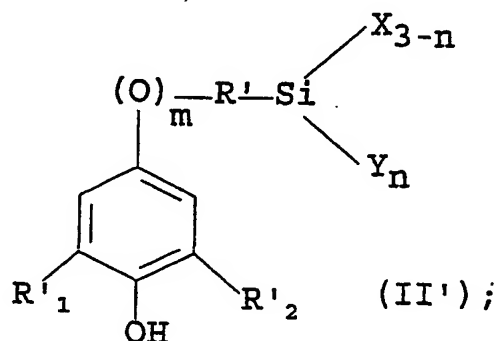


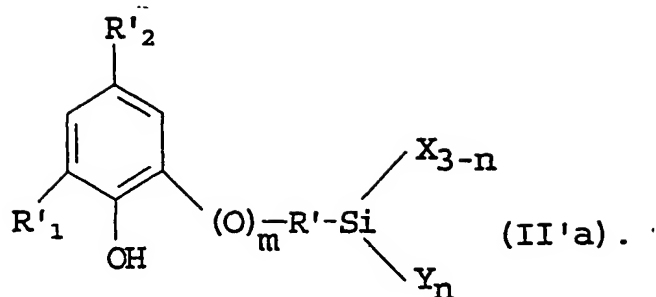
- 20       -    when n is 4, X represents a group having general formula (XX) wherein Y is bound to R<sub>2</sub>' and R<sub>2</sub>' represents a C<sub>4</sub>-C<sub>10</sub> alkane-tetrayl group.
- 25

5. The stabilizing mixtures for organic polymers according to claim 1, wherein the compounds belonging to the group of sterically hindered phenols (c) consist of reactive antioxidant compounds containing a sterically hindered phenol group having general formula (I') or (I'a):



- and more specifically, of reactive antioxidant compounds containing sterically hindered phenol groups having general formula (I') and (I'a) selected from those having general formula (II') or (II'a):

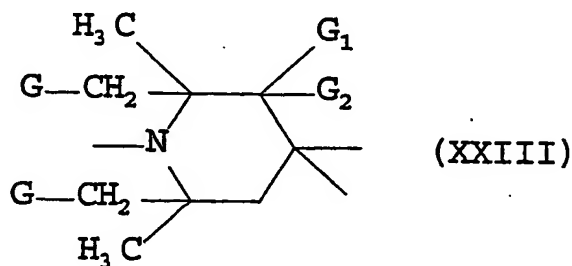




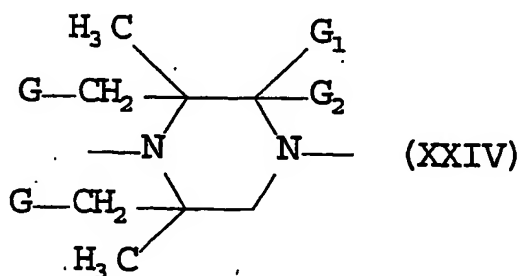
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6. The stabilizing mixtures for organic polymers according to claim 1, wherein the compounds belonging to the group of sterically hindered amines (d) are selected from those comprising at least one group having general formula (XXIII) or (XXIV):

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wherein:

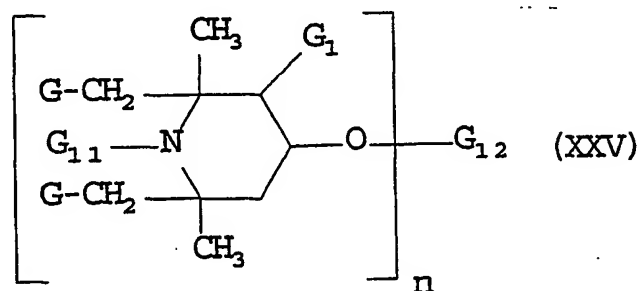
- G represents a hydrogen atom; or a methyl group;
- G<sub>1</sub> and G<sub>2</sub>, the same or different, represent a

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represent an oxygen atom.

7. The stabilizing mixtures for organic polymers according to claim 1, wherein the compounds belonging to the group of sterically hindered amines (d) are selected from compounds (a')-(h') which comprise at least one group having general formula (XXIII) or (XXIV) according to claim 6.
8. The stabilizing mixtures for organic polymers according to claim 7, wherein the compounds belonging to the group of sterically hindered amines (d) are selected from compounds (a') having general formula (XXV):

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wherein n is a number ranging from 1 to 4, extremes included; G and G<sub>1</sub>, independently, represent a hydrogen atom or a methyl; G<sub>11</sub> represents a hydrogen atom, O, a hydroxyl group, an NO group, a -CH<sub>2</sub>CN group, a C<sub>1</sub>-C<sub>18</sub> alkyl group, a C<sub>3</sub>-C<sub>8</sub> alkenyl group, a C<sub>3</sub>-C<sub>8</sub> alkynyl group, a C<sub>7</sub>-C<sub>12</sub> arylalkyl group, a C<sub>1</sub>-C<sub>18</sub> alkoxy group, a C<sub>5</sub>-C<sub>8</sub> cycloalkoxy group, a C<sub>7</sub>-C<sub>9</sub> phenylalkoxy group, a C<sub>1</sub>-C<sub>8</sub> alkanoyl group, a C<sub>3</sub>-C<sub>5</sub>



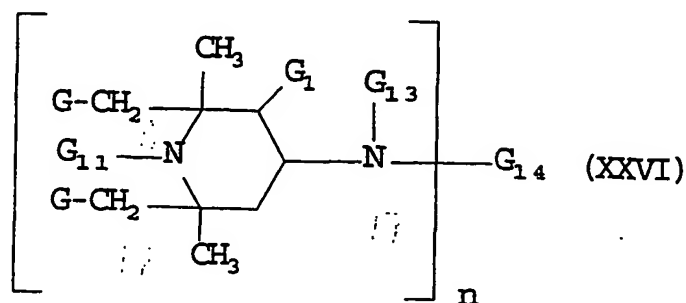
alkenoyl group, a C<sub>1</sub>-C<sub>18</sub> alkanoyloxyl group, a benzyloxyl group, a glycidyl group, an OG<sub>11</sub>' group, wherein G<sub>11</sub>' represents a linear or branched C<sub>1</sub>-C<sub>10</sub> alkyl group; a -CH<sub>2</sub>CH(OH)-Z group wherein Z represents a hydrogen atom, a methyl, or a phenyl, G<sub>11</sub> preferably being hydrogen, a C<sub>1</sub>-C<sub>4</sub> alkyl group, an allyl, a benzyl, an acetyl or an acryloyl; G<sub>12</sub>, when n is 1, represents a hydrogen atom, a C<sub>1</sub>-C<sub>18</sub> alkyl group, said alkyl group optionally containing one or more oxygen atoms, a cyanoethyl group, a benzyl, a glycidyl group, a monovalent radical of a carboxylic acid, of a carbamic acid or of an acid containing phosphorous, aliphatic, cycloaliphatic or arylaliphatic, unsaturated or aromatic, or a monovalent silyl radical, preferably a radical of an aliphatic carboxylic acid having from 2 to 18 carbon atoms, of a cycloaliphatic carboxylic acid having from 7 to 15 carbon atoms, of an  $\alpha,\beta$ -unsaturated carboxylic acid having from 3 to 5 carbon atoms, of an aromatic carboxylic acid having from 7 to 15 carbon atoms, said carboxylic acids optionally substituted in the aliphatic, cycloaliphatic or aromatic part, with 1-3 -COOZ<sub>12</sub> groups, wherein Z<sub>12</sub> represents a hydrogen atom, a C<sub>1</sub>-C<sub>20</sub> alkyl group, a C<sub>3</sub>-C<sub>12</sub> alkenyl group, a C<sub>5</sub>-C<sub>7</sub> cycloalkyl group, a phenyl or a benzyl; G<sub>12</sub>,

when n is 2, represents a C<sub>2</sub>-C<sub>12</sub> alkylene group, a C<sub>4</sub>-C<sub>12</sub> alkenylene group, a xylylene group, a divalent radical of a dicarboxylic acid, of a dicarbamic acid or of an acid containing phosphorous, aliphatic, cycloaliphatic, arylaliphatic or aromatic, or a divalent silyl radical, preferably a radical of an aliphatic dicarboxylic acid having from 2 to 36 carbon atoms, of a cycloaliphatic or aromatic dicarboxylic acid having from 8 to 14 carbon atoms, of an aliphatic, cycloaliphatic or aromatic dicarbamic acid, having from 8 to 14 carbon atoms, said dicarboxylic acids optionally substituted in the aliphatic, cycloaliphatic or aromatic part, with 1 or 2 -COOZ<sub>12</sub> groups wherein Z<sub>12</sub> has the same meanings defined above; G<sub>12</sub>, when n is 3, represents a trivalent radical of an aliphatic, cycloaliphatic or aromatic tricarboxylic acid, optionally substituted in the aliphatic, cycloaliphatic or aromatic part, with a -COOZ<sub>12</sub> group wherein Z<sub>12</sub> has the same meanings defined above, or of an aromatic tricarbamic acid or of an acid containing phosphorous, or it represents a trivalent silyl radical; G<sub>12</sub>, when n is 4, is a tetravalent radical of an aliphatic, cycloaliphatic or aromatic tetracarboxylic acid.

9. The stabilizing mixtures according to claim 7,

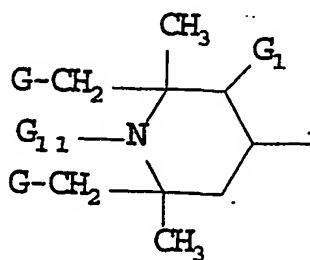
wherein the compounds belonging to the group of sterically hindered amines (d) are selected from compounds (b') having general formula (XXVI):

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wherein n is 1 or 2; G, G<sub>1</sub> and G<sub>11</sub> have the same meanings described above under claim 8; G<sub>13</sub> represents a hydrogen atom, a C<sub>1</sub>-C<sub>12</sub> alkyl group, a C<sub>2</sub>-C<sub>5</sub> hydroxyalkyl group, a C<sub>5</sub>-C<sub>7</sub> cycloalkyl group, a C<sub>7</sub>-C<sub>8</sub> arylalkyl group, a C<sub>2</sub>-C<sub>18</sub> alkanoyl group, a C<sub>3</sub>-C<sub>5</sub> alkenoyl group, a benzoyl group, or a group having the following general formula:

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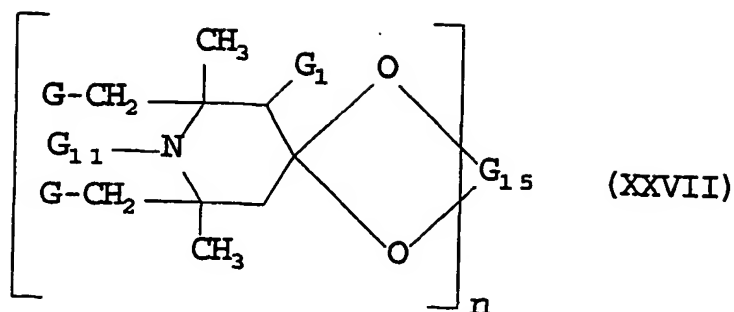
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wherein G, G<sub>1</sub>, G<sub>11</sub> have the same meanings defined above under claim 8; G<sub>14</sub>, when n is 1, represents a hydrogen atom, a C<sub>1</sub>-C<sub>18</sub> alkyl group, a C<sub>3</sub>-C<sub>8</sub> alkenyl group, a C<sub>5</sub>-C<sub>7</sub> cycloalkyl group, a C<sub>1</sub>-C<sub>4</sub> alkyl group substituted with a hydroxyl group, with a cyano

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group, with an alkoxycarbonyl group or with a carbamide group, a glycidyl group, a group having the formula  $-\text{CH}_2-\text{CH}(\text{OH})-\text{Z}$  or a group having the formula  $-\text{CONH}-\text{Z}$  wherein Z represents a hydrogen atom, a methyl or a phenyl;  $G_{14}$ , when n is 2, represents a  $\text{C}_2-\text{C}_{12}$  alkylene group, a  $\text{C}_6-\text{C}_{12}$  arylene group, a xylylene group, a group of formula  $-\text{CH}_2-\text{CH}(\text{OH})-\text{CH}_2-$  or a group having the formula  $-\text{CH}_2-\text{CH}(\text{OH})-\text{CH}_2-\text{O}-\text{D}-\text{O}-$  wherein D represents a  $\text{C}_2-\text{C}_{10}$  alkylene group, a  $\text{C}_6-\text{C}_{15}$  arylene group, a  $\text{C}_6-\text{C}_{12}$  cycloalkylene group; or, on the condition that  $G_{13}$  does not represent an alkanoyl group, an alkenoyl group or a benzoyl group,  $G_{14}$  can also represent a 1-oxo- $(\text{C}_2-\text{C}_{12})$ -alkylene group, a divalent radical of a dicarboxylic acid or of a dicarbamic acid, aliphatic, cycloaliphatic or aromatic, or also a  $-\text{CO}-$  group; or, when n is 1,  $G_{13}$  and  $G_{14}$  considered jointly, can also represent a divalent radical of a 1,2- or 1,3-dicarboxylic acid, aliphatic, cycloaliphatic or aromatic.

10. The stabilizing mixtures according to claim 7, wherein the compounds belonging to the group of sterically hindered amines (d) are selected from compounds (c') having general formula (XXVII):



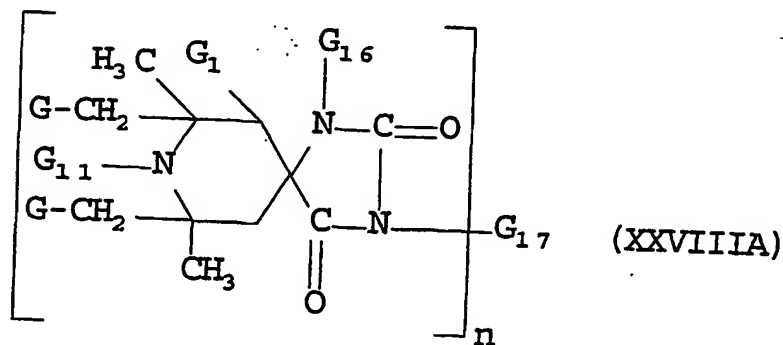
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wherein  $n$  is 1 or 2;  $G$ ,  $G_1$  and  $G_{11}$  have the same meanings described above under claim 8;  $G_{15}$ , when  $n$  is 1, represents a  $C_2$ - $C_8$  alkylene or hydroxyalkylene group, or a  $C_4$ - $C_{22}$  acyloxyalkylene group, and, when  $n$  is 2, it is a  $(-CH_2)_2C(CH_2-)_2$  group.

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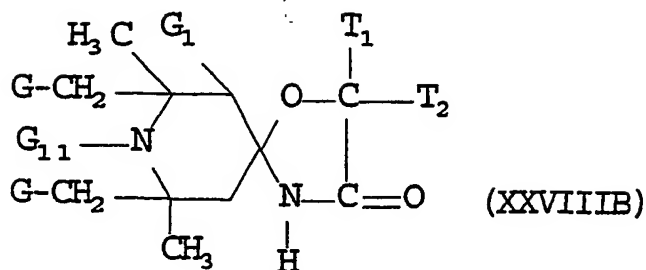
11. The stabilizing mixtures according to claim 7, wherein the compounds belonging to the group of sterically hindered amines (d) are selected from compounds (d') having general formula (XXVIII A), (XXVIII B) and (XXVIII C), the compounds having general formula (XXVIII C) being preferred:

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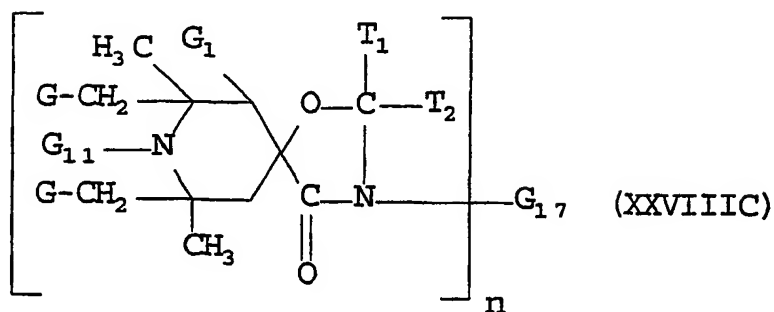


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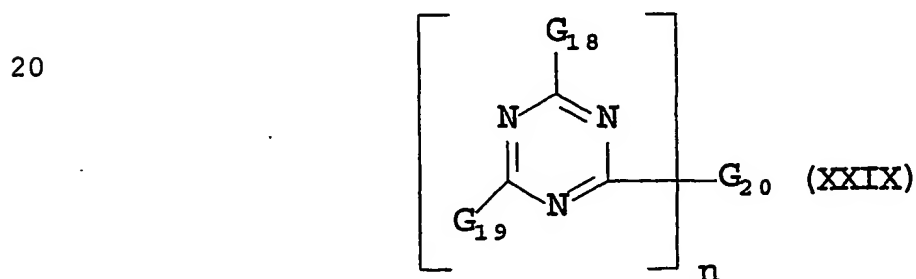
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wherein  $n$  is 1 or 2,  $G$ ,  $G_1$  and  $G_{11}$  have the same meanings defined above under claim 8;  $G_{16}$  represents a hydrogen atom, a  $C_1$ - $C_{12}$  alkyl group, an allyl group, a benzyl, a glycidyl group or a  $C_2$ - $C_6$  alkoxyalkyl group;  $G_{17}$ , when  $n$  is 1, represents a hydrogen atom, a  $C_1$ - $C_{12}$  alkyl group, a  $C_3$ - $C_5$  alkenyl group, a  $C_7$ - $C_9$  arylalkyl group, a  $C_5$ - $C_7$  cycloalkyl group, a  $C_2$ - $C_4$  hydroxyalkyl group, a  $C_2$ - $C_6$  alkoxyalkyl group, a  $C_6$ - $C_{10}$  aryl group, a glycidyl group, or a group having the formula  $-(\text{CH}_2)_p\text{-COO-Q}$  or  $-(\text{CH}_2)_p\text{-O-CO-Q}$  wherein  $p$  is 1 or 2 and  $Q$  represents a  $C_1$ - $C_4$  alkyl group or a phenyl;  $G_{17}$ , when  $n$  is 2, represents a  $C_2$ - $C_{12}$  alkylene group, a  $C_4$ - $C_{12}$  alkenylene group, a  $C_6$ - $C_{12}$  arylene group, a group having the

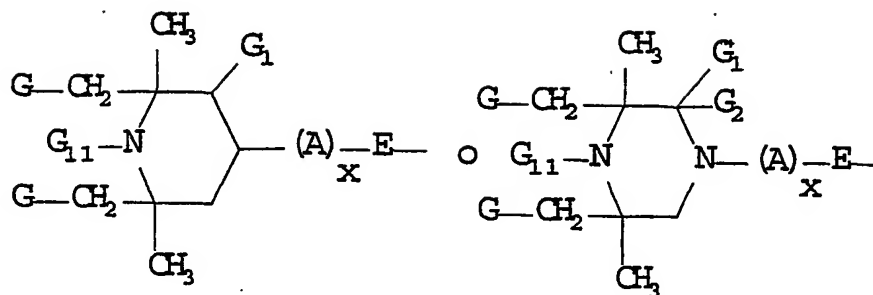
formula:  $-\text{CH}_2-\text{CH}(\text{OH})-\text{CH}_2-\text{O}-\text{D}-\text{O}-\text{CH}_2-\text{CH}(\text{OH})-\text{CH}_2-$  wherein D represents a  $\text{C}_2-\text{C}_{10}$  alkylene group, a  $\text{C}_6-\text{C}_{15}$  arylene group, a  $\text{C}_6-\text{C}_{12}$  cycloalkylene group, or a group having the formula:

5  $-\text{CH}_2\text{CH}(\text{OZ}')\text{CH}_2-(\text{OCH}_2-\text{CH}(\text{OZ}')\text{CH}_2)_2$  wherein  $\text{Z}'$  represents a hydrogen atom, a  $\text{C}_1-\text{C}_{18}$  alkyl group, an allyl, a benzyl, or a  $\text{C}_2-\text{C}_{12}$  alkanoyl group or a benzoyl;  $\text{T}_1$  and  $\text{T}_2$  each independently represent a hydrogen atom, a  $\text{C}_1-\text{C}_{18}$  alkyl group, a  $\text{C}_6-\text{C}_{10}$  aryl group, a  $\text{C}_7-\text{C}_9$  arylalkyl group, said groups optionally substituted with a halogen atom or with a  $\text{C}_1-\text{C}_4$  alkyl group; or  $\text{T}_1$  and  $\text{T}_2$  considered jointly with the carbon atom to which they are bound, form a  $\text{C}_5-\text{C}_{14}$  cycloalkane ring.

15 12. The stabilizing mixtures according to claim 7, wherein the compounds belonging to the group of sterically hindered amines (d) are selected from compounds (e') having general formula (XXIX):



25 wherein  $n$  is 1 or 2, and  $\text{G}_{18}$  represents a group having one of the following formulae:

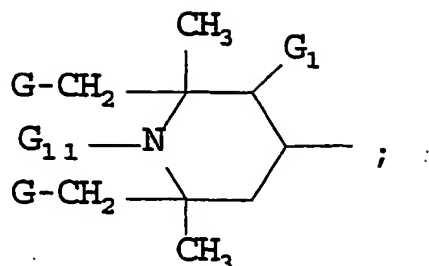


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wherein  $G$  and  $G_{11}$  have the same meanings defined above under claim 8;  $G_1$  and  $G_2$  represent a hydrogen atom, a methyl, or, considered jointly, they form a substituent  $=O$ ,  $E$  represents  $-O-$  or  $-NG_{13}-$ ;  $A$  represents a  $C_2-C_6$  alkylene group or a  $-(CH_2)_3-O-$  group;  $x$  is 0 or 1;  $G_{13}$  represents a hydrogen atom, a  $C_1-C_{12}$  alkyl group, a  $C_2-C_5$  hydroxyalkyl group, a  $C_5-C_7$  cycloalkyl group;  $G_{19}$  has the same meanings as  $G_{18}$  or it represents one of the following groups:  $-NG_{21}G_{22}$ ,  $-OG_{23}$ ,  $-NHCH_2OG_{23}$ , or  $-N(CH_2OG_{23})_2$ ;  $G_{20}$ , when  $n$  is 1, has the same meanings as  $G_{18}$ , or  $G_{19}$ , if  $n$  is 2, represents an  $-E-B-E-$  group wherein  $B$  represents a  $C_2-C_8$  alkylene group optionally interrupted by 1 or 2  $-N(G_{21})-$  groups;  $G_{21}$  represents a  $C_1-C_{12}$  alkyl group, a cyclohexyl group, a benzyl, a  $C_1-C_4$  hydroxyalkyl group, or a group having the following general formula:

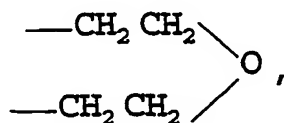
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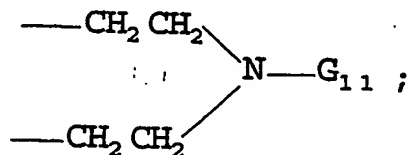


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$G_{22}$  represents a  $C_1$ - $C_{12}$  alkyl group, a cyclohexyl group, a benzyl, a  $C_1$ - $C_4$  hydroxyalkyl group;  $G_{23}$  represents a hydrogen atom, a  $C_1$ - $C_{12}$  alkyl group, a phenyl, or,  $G_{21}$  and  $G_{22}$ , considered jointly, represent a  $C_4$ - $C_5$  alkylene or oxyalkylene group, such as:

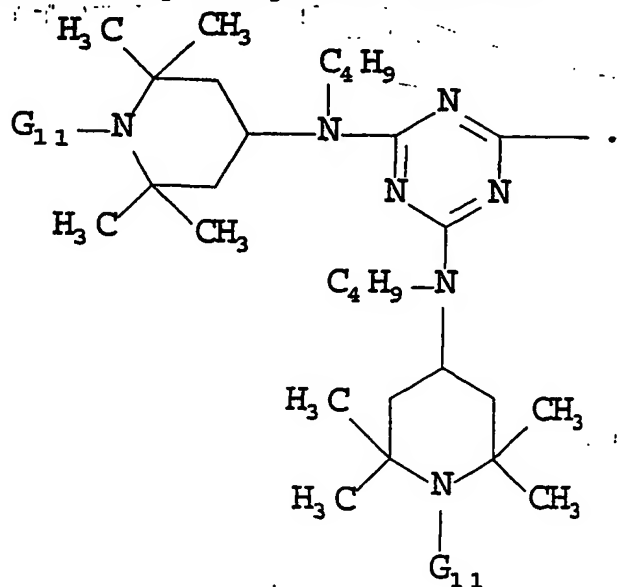


or a group having the formula:



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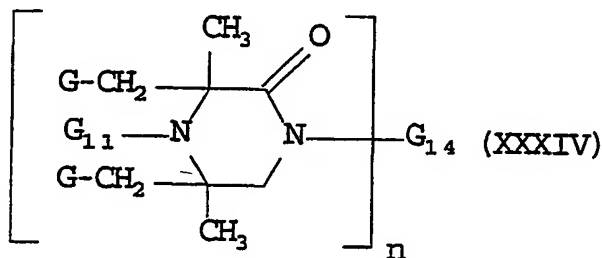
$G_{21}$  is a group having the general formula:



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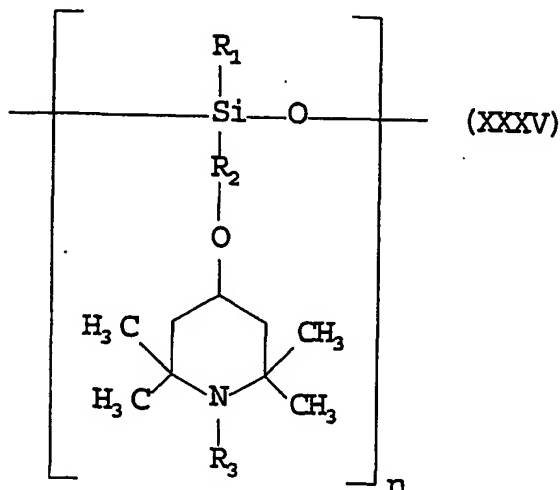
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13. The stabilizing mixtures according to claim 7,  
 wherein the compounds belonging to the group of  
 sterically hindered amines (d) are selected from  
 5 oligomeric or polymeric compounds (f') whose recur-  
 rent structural unit contains a 2,2,6,6-tetra-  
 methylpiperidine radical, in particular polyesters,  
 polyethers, polyamides, polyamines, polyurethanes,  
 polyureas, polyaminotriazines, poly(meth)acrylates,  
 10 poly(meth)acrylamides, and their copolymers contain-  
 ing said radical.
14. The stabilizing mixtures according to claim 7,  
 wherein the compounds belonging to the group of  
 sterically hindered amines (d) are selected from  
 15 compounds (g') having general formula (XXXIV):



20 wherein n is 1 or 2, G and G<sub>11</sub> have the same mean-  
 ings defined above under claim 8 and G<sub>14</sub> has the  
 same meanings described above under claim 9, on the  
 condition that G<sub>14</sub> can never represent the group  
 25 -CONH-Z or the group -CH<sub>2</sub>-CH(OH)-CH<sub>2</sub>-O-D-O.

15. The stabilizing mixtures according to claim 7, wherein the compounds belonging to the group of sterically hindered amines (d) are selected from compounds (h') having general formula (XXXV):



wherein  $R_1$  represents a  $C_1$ - $C_{10}$  alkyl group, a  $C_5$ - $C_{12}$  cycloalkyl group optionally substituted with a  $C_1$ - $C_4$  alkyl group, a phenyl optionally substituted with a  $C_1$ - $C_{10}$  alkyl group;  $R_2$  represents a  $C_3$ - $C_{10}$  alkylene group;  $R_3$  represents a hydrogen atom, a  $C_1$ - $C_8$  alkyl group,  $O$ , a  $-CH_2CN$  group, a  $C_3$ - $C_6$  alkenyl group, a  $C_7$ - $C_9$  phenylalkyl group optionally substituted in the phenyl radical with a  $C_1$ - $C_4$  alkyl group, a  $C_1$ - $C_8$  acyl group, an  $-OR'_3$  group wherein  $R'_3$  represents a  $C_1$ - $C_{10}$  alkyl group; and  $n$  is a number ranging from 1 to 50, extremes included.

16. Polymeric compositions containing an organic polymer and an effective quantity of one of the stabilizing

mixtures according to any of the previous claims.

17. The polymeric compositions according to claim 16, wherein the stabilizing mixtures are used in a combination with other stabilizers.

5 18. End-products obtained from the processing of the polymeric compositions according to claim 16 or 17.

19. A method for stabilizing organic polymers against degradation caused by oxygen, heat and/or light, which comprises the addition or application to said  
10 organic polymers of the stabilizing mixtures according to any of the claims from 1 to 15.

20. The use of 3-pyrazolidinones having general formula (I) or (II) according to claim 2, as process stabilizers for organic polymers.

15 21. Polymeric compositions containing an organic polymer and an effective quantity of at least one 3-pyrazolidinone having general formula (I) or (II) according to claim 2.

22. The polymeric compositions according to claim 21,  
20 wherein the 3-pyrazolidinones having general formula (I) or (II) are used in a combination with other stabilizers.

23. End-products obtained from the processing of the polymeric compositions according to claim 21 or 22.

## INTERNATIONAL SEARCH REPORT

Inter. Application No  
PCT/EP 03368

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C08K5/00 C08K5/3445 C08K5/49 C08K5/13 C08K5/54  
C08K5/3435

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
IPC 7 C08K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Category * (Cited documents with indication, where appropriate, of the relevant passages)	Relevant to claim No.
X	US 5 457 143 A (SCRIMA ROBERTO ET AL) 10 October 1995 (1995-10-10)  claims 1.14 figures 3B.11A, IIB, IIC, XC, XIA, XIIA column 39, line 15, 16	1-4, 6-12, 14-23
Y		5
Y	US 4 889 375 A (GRECO ALBERTO ET AL) 19 December 1989 (1989-12-19) cited in the application claim 1	5
X	GB 2 328 210 A (CLARIANT GMBH) 17 February 1999 (1999-02-17) claims 1.10, 17, 32, 34, 40 figures A4, A7, A8, B1, B7	1, 3, 6-8, 11-20

☐ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

## \* Special categories of cited documents

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Date of the actual completion of the international search

24 July 2001

Date of mailing of the international search report

01/08/2001

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## INTERNATIONAL SEARCH REPORT

Information on patent family members

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